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"Low Temperature and Neutron Physics Studies"

Abstract

Experimental studies on the diamagnetic scattering of neutrons by bismuth and copper crystals have continued. These studies are designed to furnish information on the diamagnetization density distribution around atom centers in the crystal and to search for the presence of oscillatory neutron scattering by the Landau levels associated with the conduction electrons. Other studies being performed in the laboratory include (a) determination of the nuclear spin state scattering amplitudes of Li^7 by polarized neutron scattering from nuclearly polarized crystals, (b) characterization of the phase transformation and superconducting transition in ZrV_2 and (c) phonon instabilities associated with phase and superconducting transitions in the layered compounds 2H-NbSe_2 and 2H-TaSe_2 .

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Project Research Activities

(A) Diamagnetic Scattering of Neutrons by Bismuth

C. G. Shull, W. C. Koehler (ORNL) and R. M. Moon (ORNL)

Our last project report dated August 31, 1973 has outlined the information on the diamagnetization density distribution obtained from polarized neutron scattering by bismuth. The early MIT-procured data were all collected with high magnetic field application along the $[1\bar{1}0]$ rhombohedral axis, i. e., perpendicular to the trigonal axis, and upon Fourier transformation, these yielded projection maps of the diamagnetization density onto a plane perpendicular to this axis. The maps indicated a very pronounced spatial asymmetry of the density around nuclear sites, with inverted asymmetry sense for the two unique bismuth atom sites. It is thought that this spatial asymmetry reflects that of the effective mass of the outermost 6s-6p electrons and such determination would be important in correlation with the effective mass distribution in reciprocal space seen by cyclotron resonance.

Further experiments with field application in the two other orthogonal directions were considered to be very useful and accordingly, arrangements were made with the ORNL group for a continuation of the study at the much higher intensity HFIR reactor. During January, 1974, some of the earlier studied MIT reflections were repeated at ORNL with satisfactory agreement and then other crystal specimens were studied with field application along a different direction. After studying several of the new reflections, which also showed strong suggestion of asymmetry, it was discovered that one of the reflections, (206) in orthorhombic description, exhibited a different polarization ratio upon 180° rotation thereby becoming the $(\bar{2}0\bar{6})$ reflection. This back face-front face reflection difference seemed somewhat erratic and non-reproducible and in some measurements was seen to invert sign. The significance of this, if it were a real effect and not of experiment origin, was immediately recognized since it would imply that the magnetic structure was non-centrosymmetric whereas in the early Fourier transformation it had been assumed that the magnetic structure would remain centrosymmetric, as is the crystallographic structure, upon field application along a unique axis as suggested by fundamental theoretical arguments. The development of a non-centrosymmetric magnetic structure would introduce imaginary terms in the magnetic structure factor and these in coherence with imaginary terms in the nuclear scattering could conceivably account for the observed back face-front face effects. We were suspicious from the beginning that the effect was of experiment origin and it was obviously of importance that further identification experiments be made. Several quick experiments were performed before the ORNL facilities had to be released for other work which were inconclusive in identifying the effect.

We were suspicious at the time that the observations might be plagued by simultaneous reflection effects (Renninger scattering) occurring in the Bragg scattering and during February and March, work proceeded at MIT in calculating "window wavelength" areas where this extraneous scattering could not exist. Additionally the specimen plates and pillars were given compression treatment to open up the mosaic structure thereby making the experiment less sensitive to the neutron ray

configuration in passage through the front face-back face geometry. With these specimen and technique improvements, a second limited period of data collection was scheduled at ORNL at the end of March, and these data only confirmed and made more definite the presence of a real difference. Further analysis was called for and additional experimentation at ORNL was postponed until the beginning of the summer period.

Coincident with these ORNL developments, experiments on very pure copper crystals at MIT with similar technique also showed very pronounced back face-front face differences and there was strong suspicion that the ORNL bismuth observations and the MIT copper results were related. Further work at MIT showed a dependence of the magnitude of the difference to be related to the neutron beam size and after searching for the mechanism behind this it was felt that the experiment might be reflecting the presence of oscillatory DeHaas-vanAlphen magnetization. This could arise because of the variation of the magnetic field acting at different parts of the specimen crystal (both the MIT and ORNL magnets have gradients due to unsymmetrical coil design) and in turning the crystal through 180° the effective center of gravity for scattering could be altered. One last experiment was performed at MIT before the reactor was shut down in May for rebuilding, which suggested with limited data, that this was the case. The superconducting magnet field was scanned with small increments of field (no change in crystal orientation) and changes (including inversion) of the polarization ratio somewhat beyond statistical error were observed. Further study of this at ORNL with high intensity was then scheduled for June 1974.

(B) Search for deHaas-vanAlphen Neutron Scattering by Copper

C. G. Shull, W. C. Koehler (ORNL) and R. M. Moon (ORNL)

Cyclotron resonance experiments have been remarkably effective in the determination of the Fermi surface configuration of solids and one of the simplest of such topographs is that of the noble face-centered metals, in particular that of copper. Here the surface configuration is that of spherical surfaces centered at reciprocal lattice points connected by small neck areas. Cyclotron orbits of the conduction electrons around these neck areas are excited by magnetic field application perpendicular to (111) planes in the crystal and since these orbits are quantized (Landau levels) there results an oscillatory variation of the diamagnetic susceptibility as the magnetic field is changed. As described above, some of the MIT neutron scattering observations gave strong indication that this oscillatory variation of the susceptibility was being sensed by polarized neutron scattering and a series of carefully controlled experiments were performed at ORNL. With an applied field of 57.2 KG in the [211] direction characteristic of the neutron scattering orientation, one calculates the wavelength of this field oscillation to be 117 G. Since the field gradients were known within the ORNL magnet, the neutron beam area was reduced so that the maximum field range experienced by the illuminated crystal was 40 G and the field was scanned in increments of 20 to 100 G. Polarization ratio measurements were made at each field position with the crystal being held at 4.2 K and these showed no evidence of oscillatory behavior well within the statistical error of the measurements in contradiction to the implication of the earlier MIT observations. A further

search for the oscillation structure was carried out on the (220) Bragg reflection with field application along the $[1\bar{1}1]$ axis for which the field periodicity is calculated to be 147 G. This was performed at the reduced temperature of 1.83 K to enhance the amplitude of the effect and again no oscillation in the polarized neutron scattering was seen. Thus our copper crystals certainly do not exhibit the deHaas-van Alphen oscillation in polarized beam scattering and we are ignorant at the time of writing of the origin of the MIT-ORNL differences.

In recapitulation, our interest in the copper studies had been generated as a possible explanation of the anomalous front face-back face reflection effects that had been uncovered in the bismuth diamagnetic scattering study but it now seems that this is unlikely. It may be that the failure to see the oscillatory behavior in our copper crystals is to be associated with their mosaicity since they had been given compression treatment so as to open up the mosaic structure for extinction purposes. Perfect crystals of copper are available and consideration of further experiments will be given. One may well ask why one could hope to sense such large-orbiting conduction electrons in a Bragg reflection experiment and the answer comes in that there is considerable theoretical suspicion that they may well possess considerable d-character.

(C) Polarized Neutron Scattering by Nuclearly-Polarized Li^7F

D. Frank and C. G. Shull

Coherent Bragg scattering of polarized neutrons from a crystal is in general modified if the nuclei become partially polarized and studies of this have been carried out on isotopically pure Li^7F crystals. This arises because of the spin dependence of the nuclear force interaction and the interaction is characterized by the introduction of the two fundamental spin state scattering amplitudes. These fundamental amplitudes are known for only a few nuclei since their determination requires difficult experimentation and specialized technique. There seems to be interest among nuclear theorists in having such information and the case of Li^7 is a useful one because (a) of its relative simplicity and (b) the absolute sign of the coherent scattering amplitude for unpolarized nuclei is anomalous relative to that of normal nuclear scattering.

The experiments, serving as a basis for Mr. Frank's Ph.D. thesis, have been performed with thin crystal sheets of Li^7F held in a high magnetic field of 60 KG at temperatures as low as 1.2 K. In the interpretation of the Bragg intensity change with neutron polarization inversion, it is necessary to understand quantitatively the degree of extinction in the Bragg reflection and much effort has gone into this determination as mentioned in earlier progress reports. The temperature dependence of the polarization effects has been studied in two reflections, (200) and (400) and these have individually yielded values for the spin state amplitudes which agree within experimental error. Since the start of these experiments, other experimenters at Paris have made an independent determination of these amplitudes by a different technique (a resonance spin precession method) and our values seem reasonably in agreement with these. Final values await a full interpretation of the various corrections and their uncertainties that enter into the evaluation.

(D) Phase Transformation and Superconductivity of ZrV_2

D. Moncton and W. Dobbins

X-ray experiments on polycrystalline ZrV_2 were carried out in order to determine the details of the atomic shifts involved in the martensitic transformation at $T_M = 120\text{ K}$. If this transition proceeds via a soft-phonon mechanism, then a knowledge of the low temperature structure can be used to deduce the phonon mode involved. It was found that no distortion of the atomic sublattice occurs within an experimental error sufficiently small to justify the conclusion that the transition could involve only zone center acoustic modes. The low temperature space group is $R3m$. Large diffuse scattering develops at T_M in both X-ray and neutron scattering experiments, and it is tempting to conclude that this results from phonon softening. However, it has not been possible to arrive at a consistent interpretation of this transition, as well as a similar one in HfV_2 , within the framework of soft-mode theory. Indeed recent specific heat measurements confirmed our earlier suspicion that the transition involves a fairly large latent heat and may, therefore, be strongly first order. We plan no further measurements unless a single crystal becomes available.

During the course of work on ZrV_2 , small amounts of secondary phase were consistently present in the samples, as evidenced by additional powder diffraction peaks. Combined specific heat and X-ray diffraction measurements carried out by other workers⁽¹⁾ had suggested that this secondary phase had a higher superconducting transition temperature (8.5 K) than the majority C15 phase. Discovery of the martensitic phase transformation⁽²⁾ of the C15 phase at 120 K led to the possibility that some fraction of this phase remained untransformed and was responsible for the higher superconducting transition temperature. Similar behavior is characteristic of A15 compounds and V-Ru alloys. We have succeeded in making samples of ZrV_2 that, after grinding in air and annealing, are almost entirely "secondary phase," exhibiting no measurable C15 diffraction peaks. A powder diffraction pattern of this material showed 17 peaks indicating an FCC structure with lattice constant $a_0 = 12.16 \text{ \AA}$. Chemical analysis determined that this material contained about 3.3 wt. % oxygen in addition to an essentially stoichiometric ratio of $\text{Zr}:\text{V} \sim 1:2$. Measurements of the superconducting transition temperature show a single sharp transition at 5.3 K. Thus it is likely that the higher transition temperature measured in the specific heat studies⁽¹⁾ was due to untransformed C15 phase.

(E) Transitions in 2H-NbSe_2 and 2H-TaSe_2

D. Moncton and J. D. Axe (BNL)

Neutron scattering experiments are currently in progress at Brookhaven National Laboratory on the phase transitions in 2H-TaSe_2 ($T_M = 122\text{ K}$) and 2H-NbSe_2 ($T_M = 33.6\text{ K}$). Phonon dispersion relations have been measured at 300 K and both compounds show a strong Kohn anomaly in the

1. O. Rapp and L. J. Vieland, Phys. Lett. 36A, 369 (1971).

2. D. E. Moncton, Sol. State Comm. 13, 1779 (1973).

LA phonon branch propagating in the basal plane. The wavevector of these phonons is $\vec{q} = (1/3 - \delta) a_{100}^*$. In TaSe₂ δ is small but measurable at the transition, characterizing both the critical scattering above T_M and the superlattice Bragg scattering just below T_M . As the temperature is reduced, δ decreases continuously until it becomes zero near $T = 90$ K. When $\delta = 0$, the wavevector of the distortion is commensurate with the reciprocal lattice and a unit cell having a lattice parameter of $3a_0$ is appropriate. Measurements of the intensities of superlattice peaks in the commensurate phase agree very well with structure factors calculated using a superposition of LA phonon-like distortions with wavevectors $\vec{q} = (1/3, 0, 0)$.

In addition, the temperature dependence of the order parameter has been measured for both NbSe₂ and TaSe₂. The results in the case of NbSe₂ fit the BCS functional form, while TaSe₂ shows marked deviation. It is possible that this difference is due to phonon-induced electronic lifetime effects which become more important the higher the transition temperature. An Abrikosov-Gorkov type of gap equation has been fitted to the data giving a temperature dependent lifetime consistent with resistivity measurements.

It is now generally felt that these transitions are driven by a charge density wave instability of the coupled electron-phonon system. Indeed, an incommensurate wavevector and the strong Kohn anomaly are characteristic of such an instability.

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Thesis Prepared

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Laboratory Publications

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