Physica 136B (1986) 117–119
North-Holland, Amsterdam

HELIUM CONDENSATION OBSERVED IN SMALL ANGLE NEUTRON SCATTERING

J.W. LYNN
Department of Physics, University of Maryland, College Park, MD 20742, USA
and
National Bureau of Standards, Gaithersburg, MD 20899, USA

Small angle neutron scattering (SANS) experiments have been carried out at low temperatures on powdered samples and polycrystalline ingots. A phase transition is observed in which the helium in the sample canister, used for thermal conduction purposes, condenses on the powder particles causing a large decrease in the width of the metallurgical SANS scattering. The phase transition (at constant volume) appears to be continuous and reversible.

The small angle neutron scattering (SANS) technique has provided an important tool in elucidating the nature of the competition between magnetism and superconductivity in so-called magnetic superconductors [1]. In samples where ferromagnetism is favored, for example, an oscillatory state is found to occur with a characteristic wavelength Λ given by [2]

$$\Lambda \propto (\gamma \lambda_L)^{1/2}, \tag{1}$$

where γ is the magnetic stiffness and λ_L is the London penetration depth. However, the fundamental interaction on which most theories of magnetic superconductors are based, the screening of the magnetic (dipole) interactions by the supercurrents, has not been verified directly. Well above the magnetic phase transition where the magnetic correlations are negligible, for example, the magnetic scattering ($\chi(q)$) should be independent of the wave vector q. Physically this means that all fluctuations of the magnetization density are equally likely. Below the superconducting transition temperature, however, magnetic fluctuations with wavelengths comparable to or larger than the London penetration depth will be energetically unfavored, and hence the scattering at the corresponding wave vectors ($Q \leqq 1/\lambda_L$) should be reduced. We have attempted to observe this (small) suppression directly in a number of magnetic superconductors, including $HoMo_6S_8$, $HoMo_6Se_8$, and $(Ce_{1-x}Ho_x)Ru_2$, without success. However, in the course of these studies we have found that the helium that is often used in the sample environment for thermal conduction purposes condenses on the sample, and that this condensation can produce very large changes in the metallurgical scattering from powder specimens. These changes are qualitatively similar, but much larger than, the expected magnetic suppression effect.

The data were taken on the small angle neutron scattering spectrometer at the National Bureau of Standards Research Reactor, and the measurements were made in the high-resolution mode. The powdered samples were placed in an aluminum can which was filled with helium in a glove bag, and then the sample can was sealed and mounted in a cryostat. Fig. 1 shows the total scattering observed on the two-dimensional position-sensitive detector as a function of temperature for two samples. For $(Ce_{0.9}Ho_{0.1})Ru_2$, which has a superconducting transition temperature of 6.30 K and no transition to magnetic long range order, we observe a large decrease of the overall intensity below 1.455 K, indicating the onset of a phase transition at this temperature. Note that the transition appears to be continuous and reversible. Since the change in intensity is so large (the total intensity has decreased by ~50 000 counts, which is about 15% of the total signal), this immediately rules out the superconductive sup-

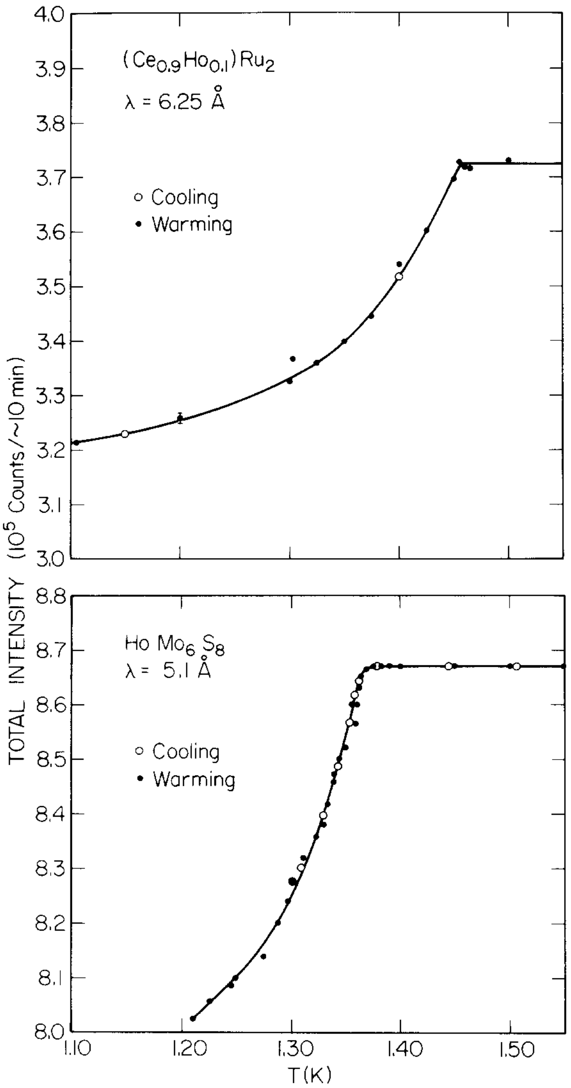

Fig. 1. Total intensity on the two-dimensional position-sensitive detector as a function of temperature for two different polycrystalline samples. The large decrease in the intensity identifies the onset of helium condensation in the sample canister.

pression effect, which should be orders of magnitude smaller, as the origin of this transition. In addition, the transition temperature of 1.455 K is unrelated to any known property of the sample. A similar transition is observed in $HoMo_6S_8$ (bottom half of fig. 1), which has a superconducting transition at 1.82 K and a magnetic/reentrant transition at about 0.7 K. The transition also appears to be continuous and reversible, and occurs at a somewhat lower temperature of 1.37 K. Again the magnitude of the effect is very large, and the transition temperature does not correspond to any known phenomenon in the sample. Finally, data on a solid ingot of $(Ce_{0.9}Ho_{0.1})Ru_2$ showed no such transition.

The change in the wavevector dependence of the scattering which is caused by the transition is shown in fig. 2 for a powder sample of $(Ce_{0.9}Ho_{0.1})Ru_2$. Here we have subtracted the scattering at 1.5 K from the data at the lowest temperature taken, 1.1 K. We see that the intensity decrease occurs at small Q, and we attribute this to the condensation of helium onto the particles. The liquid helium has the effect of smoothing the particles, and making them larger so that the scattering shifts to small Q. In this respect the intensity changes found in fig. 1 are really artificial in that there is no real change in the total intensity. Actually the intensity shifts to sufficiently small Q that it is absorbed by the beam-mask which is placed in the forward beam direction so that the detector does not saturate with counts. Measurements with an attenuator in the incident beam and the detector beam-mask removed showed that there is no

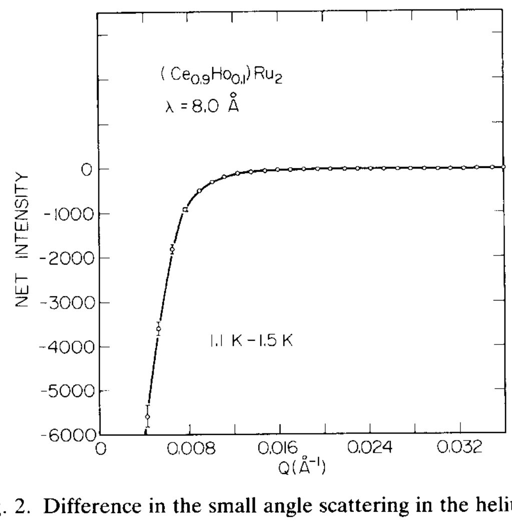

Fig. 2. Difference in the small angle scattering in the helium-condensed phase. The effect is largest at small Q. The data do not extend below 0.004 $Å^{-1}$ since this is the region where the transmitted beam is masked.

change in the sample transmission, but rather that the transmitted beam is narrower below the condensation temperature. This demonstrates directly that the profile of the scattering is narrowing, which means that the average size of the particles is increasing.

Fig. 3 shows the same kind of subtracted data for $HoMo_6S_8$ for four different temperatures. At 1.50 K, which is above the condensation temperature, there is a slight increase in scattering at small Q due to the development of some correlation of the magnetic moments. Below the condensation temperature the scattering abruptly decreases at small Q, in a continuous fashion. We believe this is due to the increase of the thickness of the helium film on the powder particles as the size of the "droplets" grows. This behavior is typical of condensation in a constant-volume environment. Of course, the gas-liquid transition is first-order in nature, and this can be directly observed if the gas is condensed at constant pressure rather than constant volume. An experiment of this type was carried out on a sample of non-magnetic YMo_6S_8 on the D-11 small angle spectrometer at the Institut Laue Langevin [3]. The sample holder in this case can be flooded with helium [4], which was found to decrease the total intensity on the detector (with a beam mask in place) by a factor of two. In this case the scattering changes because the neutron detects a different "contrast", from vacuum–sample–vacuum to helium–sample–helium. This decrease in contrast reduces the strength of the scattering (and multiple scattering). Contrast-matching is a standard technique used in many SANS experiments.

One final remark is that we did not observe any change in the scattering due to the superconducting phase transition in any of these samples, with the possible exception of $(Er_{0.16}Ho_{0.84})Rh_4B_4$ [5]. The strong small-Q scattering from these powders makes such measurements very difficult, and it is likely that the effect can only be observed unambiguously in high quality polycrystalline or single crystal samples.

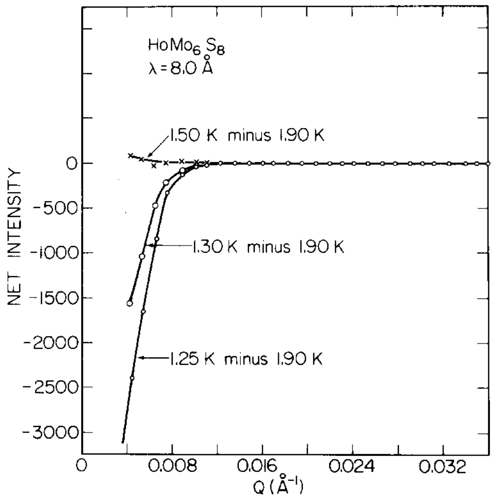

Fig. 3. Difference data for $HoMo_6S_8$. Above the condensation temperature of 1.37 K (for this sample) there is a small increase in the scattering due to development of magnetic correlations in the sample. Below 1.37 K there is a sharp decrease in the scattering at small Q, which is more pronounced with decreasing temperature.

Acknowledgements

We would like to thank C.J. Glinka and R. Pynn for helpful discussions and assistance. Research at Maryland was supported by the National Science Foundation under grant No. DMR 83-19936.

References

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[3] Experiment No. 7-01-224, in collaboration with R. Pynn, approved by the Scientific council 10/81.
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