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SINGLE CRYSTAL NEUTRON DIFFRACTION STUDY OF THE MAGNETIC STRUCTURE OF $TmNi_2B_2C$

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Abstract

Neutron diffraction techniques have been used to study the magnetic structure of single crystals of the magnetic superconductor ($T_c \cong 11K$) $TmNi_2B_2C$. We find that below approximately 1.5K the magnetic moments order in an incommensurate spin wave with propagation vector $\vec{q}_m = q_m (\vec{a}^* + \vec{b}^*)$ (or $\vec{q}_m = q_m (\vec{a}^* - \vec{b}^*)$) with $q_m = 0.094 \pm 0.001$. The spin wave is transverse with the moments aligned along the c-axis, and the observation of relatively intense higher order harmonics shows that the modulation is not purely sinusoidal but considerably squared. This incommensurate magnetic structure, which coexists with superconductivity below $T_N \cong 1.5K$, is quite different from those observed in the magnetic superconductors $HoNi_2B_2C$ and $ErNi_2B_2C$. The origin of diffraction peaks observed in scans parallel to $\vec{a}^*$ is briefly discussed.

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I. Introduction

The compounds of the recently discovered [1-4] family of rare-earth nickel boride carbides, RNi_2B_2C (where R stands for a rare-earth element) have very interesting physical properties at low temperatures. The structure [3] of these compounds is body-centered tetragonal (space group I4/mmm) and consists of R-C layers separated by Ni_2B_2 sheets; typically $a \cong 3.5\mathring{A}$ and $c \cong 3a$. Many of these compounds (R=Lu,Y, Tm, Er, Ho, Dy) are superconducting, and the highest observed [2-6] superconducting temperatures are 16.6K for the Lu, and 15.6K for the Y compound. Compounds containing magnetic rare-earth elements, such as Tm, Er, Ho and Dy, have also relatively high superconducting transition temperatures [2-7] (10.8, 10.5, 8.7 and 6.2K respectively). In the Tm, Ho and Er compounds the magnetic ordering temperature, T_N, is below the superconducting transition temperature T_c, whereas in the Dy compound $T_N > T_c$. Other compounds of this family containing magnetic rare-earth elements (Gd, Tb, Nd, Sm) are not superconducting (at least down to approximately 2K) but order magnetically at low temperatures.

The magnetic structures of the RNi_2B_2C compounds have been studied by neutron and resonant x-ray scattering techniques. A simple commensurate antiferromagnetic structure was observed [7-10] in the Dy, Nd and Sm compounds and below 4.7K in the Ho compound [11,12]. Incommensurate magnetic structures were also observed in the ordered state of many of these compounds. Of particular interest is the observation of an incommensurate modulation with propagation vector along $\vec{a}^*$ in the superconducting Er[13,14] and Ho [11,15] systems and the non-superconducting Tb[16] and Gd[17] compounds. Since the magnitude of the wave vector does not vary appreciably among these compounds (0.553,

0.585, 0.551 (just below 15K), 0.553 for Er, Ho, Tb and Gd respectively), this observation suggests that there are common Fermi surface nesting features along $\vec{a}^*$ in the RNi_2B_2C family that cause the ordering of the rare-earth moments via the RKKY mechanism. The observation[18] of soft phonon modes at wave vectors close to this incommensurate magnetic ordering wave vector in $LuNi_2B_2C$ strongly supports this conclusion. Furthermore, this result is in agreement with the results of band theoretical calculations[19] of the generalized electronic susceptibility of $LuNi_2B_2C$.

$TmNi_2B_2C$ has a superconducting transition temperature of approximately 11K. Magnetization[20] and specific heat[21] measurements show that this compound orders magnetically at approximately 1.5K. The magnetic anisotropy in this compound is different[20] from that observed in the other (Ho, Er, Dy) magnetic superconductors of this family and implies that the ordered moments in this compound are not confined in the a-b plane, as in Ho, Er and Dy, but they are along the c-axis of this tetragonal structure. In this brief paper we present the results of a neutron diffraction study of single crystals of this compound.

II. Experimental Details.

The samples used in the present experiment were single crystals depleted in the strongly neutron absorbing B^{10} nuclei[20]. A systematic search for magnetic diffraction peaks was performed at low temperatures in both the a-b and a-c planes. For this purpose we used two differently mounted single crystals. For both searches the crystal was in an He^3 cryostat with base temperature of approximately 0.35K.

The neutron diffraction experiments were performed using the H4M triple-axis spectrometer at the high flux beam reactor (HFBR) of the Brookhaven National Laboratory. Pyrolitic graphite, reflecting from the

(002) planes, was used as both monochromator and analyzer and a pyrolitic graphite filter was placed after the sample to attenuate higher-order contamination. Most measurements were performed at 14.7 meV with collimation of 40-40-40-80 minutes of arc.

III. Experimental Results and Discussion

The diffraction pattern from the crystal was determined in both the a-b and a-c crystal planes at room temperature, 4.2K and several temperatures between 1.5 and 0.35 K, the lowest temperature reached in these experiments. At temperatures above approximately 1.5K only nuclear reflections (hkl) with h+k+l=2n are observed as expected from the crystal structure of the compound. Thus, at temperature above approximately 1.5K the magnetic moments in $TmNi_2B_2C$ are not ordered.

Below approximately 1.5K additional diffraction peaks are observed in rows parallel to the [110] direction in reciprocal space (see Fig. 1). These diffraction peaks can be indexed (Fig. 1) as first and higher order satellites of the allowed (h+k+l=2n) nuclear reflections. Similar diffraction patterns were observed in scans parallel to the equivalent $[\bar{1}10]$ direction of the tetragonal lattice. Since the diffraction peaks observed in scans parallel to these two equivalent directions are of comparable intensities, it is reasonable to assume that the [110] and $[\bar{1}10]$ domains are equally populated. These observations imply that below approximately 1.5K the magnetic moments in $TmNi_2B_2C$ order in an incommensurate modulated magnetic structure with wave vectors parallel to the two equivalent [110] directions. The propagation vector obtained from the positions of the satellites is $\vec{q}_m = q_m (\vec{a}^* + \vec{b}^*)$ (or $\vec{q}_m = q_m (\vec{a}^* - \vec{b}^*)$) with $q_m = 0.094 \pm .001$ at 0.35K. No appreciable change was observed in the magnitude of the propagation vector between 0.35 and 1.5K. The

intensities of the observed satellites show that the magnetic moments are perpendicular to the scattering plane which coincides with the a-b plane of the crystal. Thus, the magnetic moments are aligned along the c-axis of the crystal, in agreement with the results of magnetization measurements.[20] These results are in very good agreement with those obtained in recent studies[22,23] of powder samples of this compound. Below 1.5K the intensity of the observed satellites increases with decreasing temperature and starts approaching saturation at 0.35K, the lowest temperature reached in the present experiments (see Fig. 2). Thus below approximately 1.5K the magnetic structure of $TmNi_2B_2C$ can be described as a transverse spin wave with propagation vector along the [110] (or equivalent [$\bar{1}$10]) direction. This transverse incommensurate modulation of the moments is not purely sinusoidal but considerably squared as shown by the observation (see Fig. 1) of relatively intense higher order harmonics.

In addition to the magnetic peaks observed in scans parallel to the [110] (or [$\bar{1}$10]) direction, weak diffraction peaks were observed in scans parallel to $\vec{a}^*$ (or $\vec{b}^*$). These peaks can be indexed as "satellites" to the nuclear reflections with wave vectors q_m a* and 2 q_m a*. An attempt to study in detail the origin of these peaks with polarized neutrons failed because of the low peak intensities (several thousand times smaller than those of the nuclear reflections) of these diffraction peaks. It should be pointed out, however, that the appearance of these weak peaks may be attributed to resolution effects ("satellites" with wave vector q_m a*) and double magnetic scattering ("satellites" with wave vector 2 q_m a*).

The most interesting result of the present experiments is that the incommensurate magnetic structure of $TmNi_2B_2C$ is quite different from that observed in the other two magnetic superconductors (Ho, Er) of the

RNi_2B_2C family that also order in an incommensurate structure. As we mentioned earlier (see Introduction) there is ample evidence that the ordering in the Ho, Er, Tb, and Gd compounds is due to a common nesting feature along $\vec{a}^*$ of the Fermi surface. It would therefore be interesting to ascertain whether the ordering in this compound may also be due to a nesting feature of the Fermi surface. If this is the case, one would expect to observe a Kohn anomaly in the phonon dispersion curves of this compound at a wave vector close to the magnetic ordering vector $\vec{q}_m$.

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Figure Captions

Fig. 1. Typical scan along the [hho] symmetry direction. The satellites of an [hho] reflection are denoted by $(\mathrm{hho})_n^{\pm}$ where the superscript + (or -) means that magnetic wave vector is added (or subtracted) from the reciprocal vector of the reflection, and the subscript n is the order of the satellite. The (out of scale) peak intensities of the (110), $(110)_1^{+}$, $(110)_1^{-}$ lines are 64319, 37701, and 26728 respectively. 8400M (monitor counts) correspond to approximately 117 sec.

Fig. 2. Temperature dependence of the $(110)_1^{-}$ satellite.

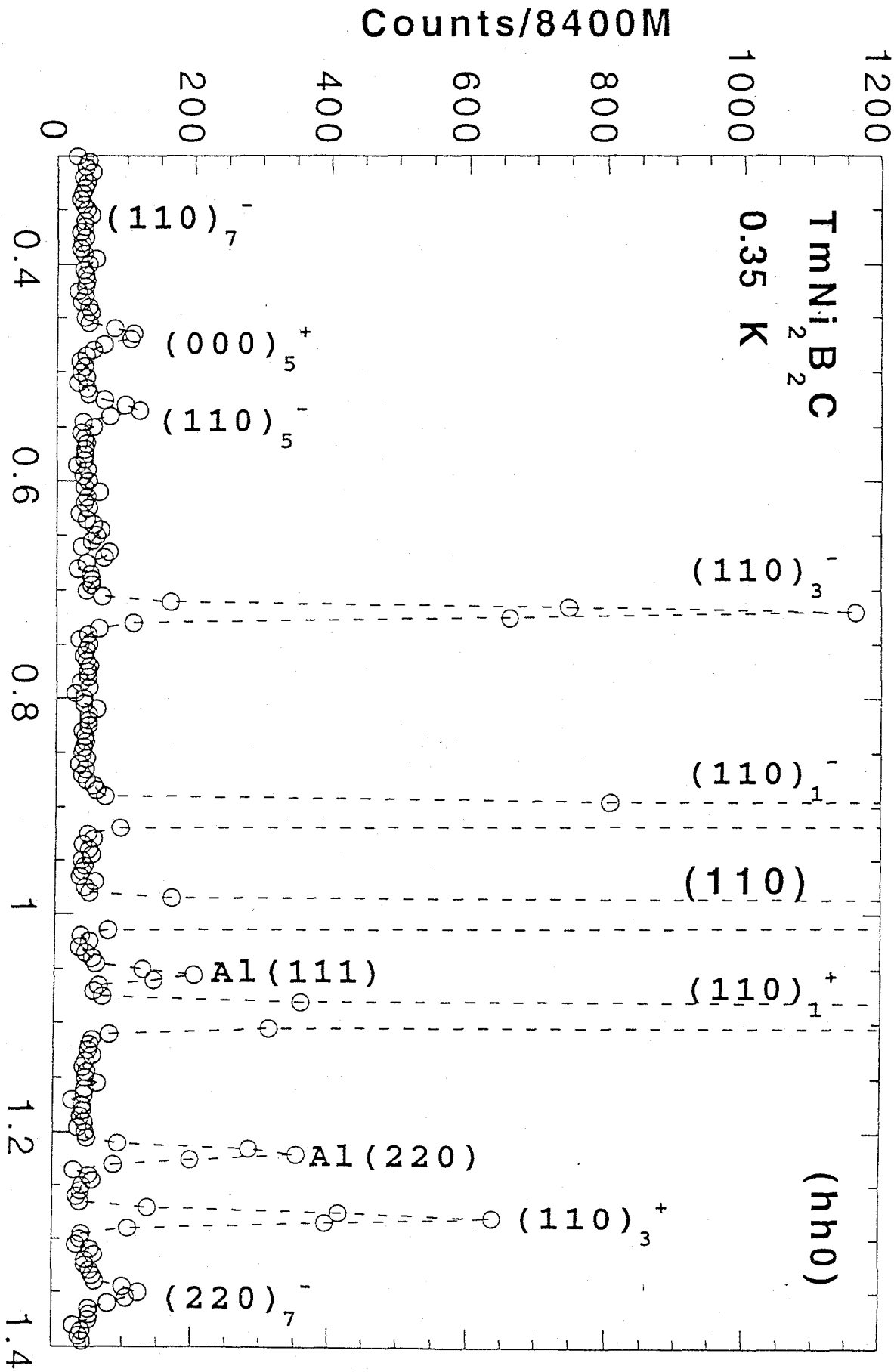
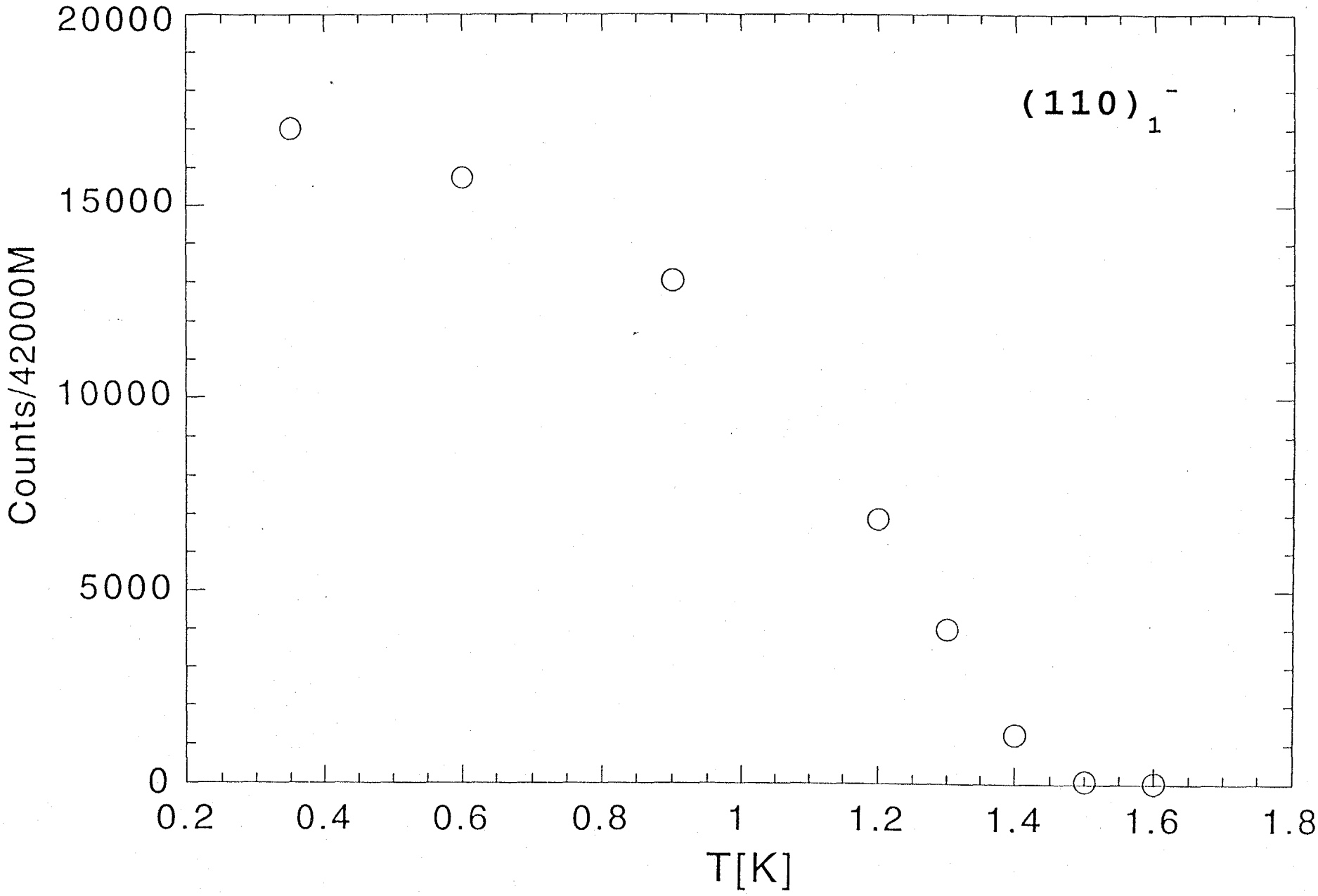


Fig. 1 B. Sternlieb J. Appl. Phys.



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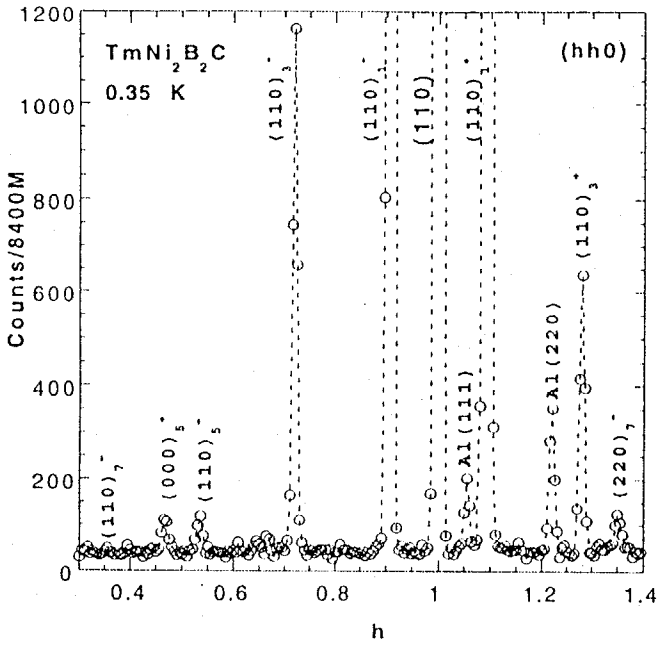

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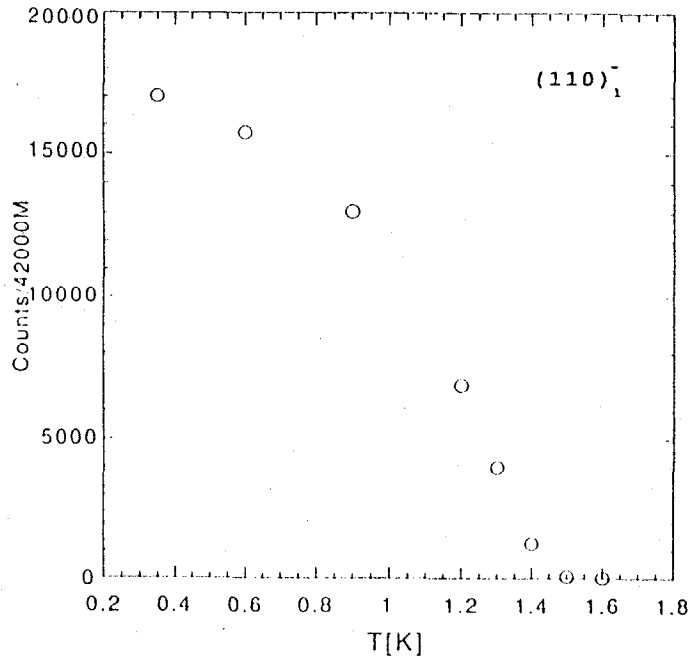

Fig.2 B. Sternlieb J. Appl. Phys.