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SPIN DYNAMICS AND MAGNETIC ORDERING IN MIXED VALENCE SYSTEMS

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ABSTRACT

Neutron scattering measurements are reported on the mixed valence compounds $Ce_{1-x}Th_x$ and TmSe. The $\chi''(Q,\omega)$ as derived from the inelastic spectra of $Ce_{0.74}Th_{0.26}$ shows a peak in the γ phase near 20.0 meV and shifts abruptly to greater than 70.0 meV at the transition to the α phase. The temperature independence of the susceptibility within the γ phase cannot be simply reconciled with the temperature dependence of the valence within the γ phase. TmSe is shown to order in a type I antiferromagnetic structure below $T_N \sim 3.2$ K. The magnetic phase diagram is understood as a successive domain reorientation and a metamagnetic phase transition for $T < 3$ K with increasing field. The mixed valence nature manifests itself in a reduced moment and a markedly altered crystal field. Another sample of TmSe with a lattice parameter implying 100% Tm^{3+} orders in a type II structure but never achieves long range order.

INTRODUCTION

Over the past several years interest in mixed valent materials has increased enormously[1,2]. Various types of experimental probes have been used in order to understand the detailed mechanisms giving rise to a non-integral valence of the magnetic ion. Unfortunately, there is no satisfactory theory to explain the experimentally observed features in these materials. In fact, in an amusing and insightful anecdote, Anderson has likened the theoretical situation to the blind man touching the elephant[3]. Depending upon where he touches, he guesses it is something quite different from an elephant, but an object known from his other experiences. In this review, we would like to report on some of our neutron scattering experiments on mixed valence materials in order to illuminate this "elephant," at least insofar as magnetic fluctuation phenomena and

DISCLAIMER

magnetic ordering are concerned. As we shall see, although neutrons provide certain very important pieces of information, we have by no means solved the mixed valent problem. Nevertheless, combining this and related work we now have a fairly complete empirical description of the static and dynamic magnetic properties of several prototypical rare earth mixed valence systems.

The two compounds we shall discuss are $Ce_{1-x}Th_x$[4] ($x = 0.26$) and TmSe[5]. The former undergoes a mixed valent transition as the temperature is decreased, from a predominantly Ce^{3+} γ phase, to a more mixed state of Ce^{3+} - Ce^{4+}, the α phase. The dynamics of this system have been studied by inelastic neutron scattering. TmSe is in the mixed valence state (typically 80% Tm^{3+} - 20% Tm^{2+}) at all temperatures and the ratio, Tm^{3+}/Tm^{2+}, appears to be determined by the growth mechanism of this material. We shall show that this material does indeed exhibit long range magnetic order which surprisingly depends upon the Tm^{3+}/Tm^{2+} ratio.

All neutron experiments were performed on a triple axis spectrometer at Brookhaven National Laboratory's high-flux beam reactor. The experimental details are given in References [4] and [5].

$Ce_{1-x}Th_x$

The measurements were made on a polycrystalline sample of $Ce_{1-x}Th_x$ with $x = 0.26$. Resistivity and susceptibility measurements on this same sample showed a first order phase transition occurring at $T_V \sim 153$ K[4]. Measurements of the lattice parameter also confirmed the first order nature. From this temperature dependence and using the linear extrapolation technique, Végard's Law, we were able to estimate the temperature dependence of the valence. This is shown in Fig. 1 where the right hand scale shows the fractional occupancy of the 4f level. It is seen that the Ce ion is in a mixed valence state at all temperatures with a valence of 3.17 at T = 300 K and a gradual increase to 3.29 at T = 153.5 K just above T_V. At T_V there is an abrupt increase in valence to 3.39 just within the α phase.

In Fig. 2, we show the imaginary part of the susceptibility, $\chi''(Q,\omega)$ as derived from the observed inelastic neutron scattering spectra. These curves were obtained after subtraction of the phonon background deduced from an identical size and shape sample of $La_{0.73}Th_{0.27}$[4]. The most obvious feature in this figure is the dramatic change of energy scale of the scattering as the crystal transforms into the α phase. The

relationship between this spin fluctuation energy and the electronic energies is not yet clear. In comparing with the behavior of the valence, we see that the change in valence between room temperature and T_V is the same as the change at T_V. If we use the notion of $E_F - E_f$ decreasing to explain this behavior then in order to reconcile the constant behavior of $\chi''(Q,\omega)$ in the γ phase with the abrupt change at T_V, one requires a highly nonlinear dependence of $E_F - E_f$ on the valence. This undoubtedly involves a complicated interplay between thermal valence excitations and quantum mechanical valence mixing. A quantitative theory which treats this problem properly would be most welcome. Related questions arise when the observed neutron intensity is placed on an absolute scale. How does the change in valence enter the neutron cross section? For a perfect paramagnet, the expression for the cross section is well known[4]. However, for a fractional valence (Fig. 1) at intermediate temperatures, it is not clear whether $n(T)$ should enter the cross section expression linearly as would be the case for an incoherent thermally activated process, or quadratically as would be the case for a coherent mixture of Ce^{3+} and Ce^{4+} wave functions. The latter seems to be more consistent with the experimental data. As we shall see below for TmSe, there may even be a crossover from a coherent to an incoherent mixed valent state.

Interesting results are also obtained for the Ce form factor. Fig. 3a shows the form factor measured by looking at the Q dependence of the scattering at an energy transfer of $\Delta E = 30.0$ meV. This is well beyond the phonon cutoff and therefore arises only from magnetic scattering. This measurement was taken just above the phase transition where there is already a significant mixing of the valence. Again, the electron is presumably spending a considerable amount of time in the conduction band and from the simple picture, one might expect to see an anomalous contribution to the form factor arising from the more extended nature of the electronic wave function. Measurements down to Q values of 0.3 $Å^{-1}$ revealed no anomalous behavior. In fact, the solid line in Fig. 3a is just the calculated atomic Ce^{3+} form factor.

Comparing the results of Fig. 2 with the bulk susceptibility measurements, we find that the energy scales are quite comparable since the latter measurements imply spin fluctuation energies of order 12 and 130 meV in the γ and α phases respectively. Since the static and the dynamic measurements are closely similar to each other, we conclude that the valence fluctuations are essentially local excitations in $Ce_{1-x}Th_x$.

TmSe

In most mixed valence systems, one of the rare earth valence states is magnetic and the other one is nonmagnetic. In this case, one might not expect long range magnetic order to exist since, as the valence fluctuates, the spins may not remember which state they should belong to. This conjecture, in fact, originates primarily from extensive empirical data, in such systems which indicate no magnetic long range order. TmSe is a mixed valence system in which both of the valence states are magnetic and as predicted by Varma[1] long range magnetic order should occur. Unfortunately, up until recently the existence of long range magnetic order in TmSe has not been clear. Early neutron diffraction experiments in a polycrystalline sample were ambiguous[6], but more recent macroscopic measurements suggested a well-defined phase transition at $T \sim 3$ K [7-11]. The situation in TmSe is even more complex because results appear to be very sample dependent[11,12]. Below we present field and temperature dependent neutron diffraction results on two samples of TmSe. Sample no. 1 was supplied by Walsh et al.[13] and corresponds to a good sample from the top of their boule. Resistivity and susceptibility measurements on adjacent pieces indicate a well-defined phase transition at ~ 3 K. The lattice parameter of this material was 5.71 Å at room temperature and using Végard's law corresponds to mixed valence of 80% $Tm^{3+}(4f^{12})$ and 20% $Tm^{2+}(4f^{13})$. The second sample, no. 2, which was grown several years ago by one of us.(E.B.) is not as well characterized but has a lattice parameter of 5.64 Å which implies almost 100% Tm^{3+}. We emphasize that these estimates are only approximate. Sample 2 also has a purplish hue in contrast to sample 1 which was gold colored. Recent chemical characterization of other TmSe samples suggest that the samples may be $\sim$ 13% Tm deficient[12]. Resistivity measurements on similar samples revealed no well-defined phase transition[12] but as we shall see below, a quasi ordering occurs near $T \sim 4.6$ K.

In sample no. 1, below 3.2 K magnetic scattering appears at all of the forbidden face centered cubic (fcc) reflections with h,k,ℓ neither all even or all odd[5]. This is consistent with a type I antiferromagnetic order, which is a tetragonal magnetic structure with spins lying in planes perpendicular to one of the cube axes. The magnetic moments within the planes are parallel, but oppositely directed in successive neighboring planes directed along the tetragonal axis (Fig. 4). By comparing the intensities of 16 magnetic superlattice reflections we conclude, unambiguously, that the antiferromagnetic

order consists of a type I structure with spins pointing along the (100) directions, perpendicular to the tetragonal axis. The magnetic form factor obtained from these measurements are shown in Fig. 3b. By comparing the magnetic intensities to the weak nuclear reflections, we deduce an ordered antiferromagnetic moment of $1.7\pm0.2\mu_B$ a value significantly lower than the $6.5\mu_B$ saturation moment estimated for an 80-20 mixture of Tm^{2+} and Tm^{3+}. It is important to note that this latter value was measured at higher temperatures via the Curie behavior of the susceptibility[7-9].

Under application of a magnetic field along a [100] direction we observed a magnetic phase diagram similar to that reported by others[8,9]. However, using neutron diffraction, we can monitor directly the behavior of the spins. We arrive at the following rather simple explanation of the phase diagram shown in Fig. 1[5]: I) Phase boundary I corresponds to an apparently second order phase transition into the antiferromagnetic state discussed above. II) This phase boundary only occurs in samples that have been cooled in a zero field and corresponds to redistribution of domains from six in a polydomain sample, to two at fields of $H \sim 2kG$. For samples that have been field-cooled, so that the minimum number of domains exist, this boundary is no longer present. III) This phase boundary at $H \sim 3kG$ corresponds to the disappearance of the antiferromagnetic intensity and an abrupt increase in the ferromagnetic intensity. At this field, the spins suddenly align themselves and a first order metamagnetic phase transition occurs. Lines I and III most likely meet at a multicritical point. The exact nature of the transitions at I and III are difficult to determine in the present experiment because of demagnetizing effects. Studies of properly shaped samples should yield interesting information on the critical phenomena in mixed valent systems. IV) Phase boundary IV has not been observed in the neutron experiments but represents points at which the bulk susceptibility is maximum. In our picture, this line represents an extension of the multicritical point into the paramagnetic phase.

The above phase diagram may be readily accounted for by assuming a simple classical spin Hamiltonian containing a Zeeman term, a cubic anisotropy, nearest neighbor(nn), and next nearest neighbor interaction(nnn)

$$\mathcal{H} = \sum_i g u_B H \cdot S_i + 5/2 B_4 \sum_i (S_i^+)^4 + (S_i^-)^4 + \sum_{nn} J_{nn} \vec{S}_i \cdot \vec{S}_m + \sum_{<nnn>} J_{nnn} \vec{S}_i \cdot \vec{S}_m \qquad (1)$$

The S_i's are considered as unit classical vectors and are assumed to point along the (100) direction. H is the magnetic field which will rotate the spins from the [100] direction towards the [010] direction (Fig.4). If α is the angle the spin makes with the [100] direction, the angle dependent energy per spin in the mean field approximation is

$$E = 4J_{nn}\cos 2\alpha - 4B_4\cos 4\alpha - gu_B H\sin\alpha \qquad (2)$$

By minimizing this with respect to α, a relation between the field and α is found.

Also within the mean field approximation

$$kT_N = -4/3J_{nn} + 2J_{nnn} \qquad (3)$$

From our data and that of Walsh et al. [13] the low temperatures boundary III occurs at 2.8 kG and $\sin\alpha\sim 0.4$ for this field. From these results we obtain

$$B_4 \simeq 4 \times 10^{-4} \text{ meV}$$

$$J_{nn} = -3 \times 10^{-3} \text{ meV} \qquad (4)$$

$$J_{nnn} = 1.3 \times 10^{-1} \text{ meV}$$

B_4 is remarkably small and markedly sample and pressure dependent. B_4 is also of opposite sign to that estimated on a point charge model and observed in related pnictide compounds [14].

The mixed valence nature of TmSe seems to manifest itself principally in the anomalously low moment and the small value and opposite sign of the crystal field parameter. We believe that the small moment is not due to crystal field effects since a moderate magnetic field should cause a saturation of the moment. Experimentally it has been observed that even fields up to 150 kG show no sign of saturation and an induced moment of only $\sim 4\mu_B$ is observed[9,11].

The microscopic origin of this low value of the moment can be represented by considering as a wave function for the mixed valence state

$$|mv\rangle = \alpha|3+\rangle + \beta|2+\rangle \qquad (5)$$

where in our sample $\alpha^2 = 0.8$ and $\beta^2 = 0.2$. The magnetic moment is then proportional to

$$\langle mv|J|mv\rangle = \alpha^2\langle 3+|J|3+\rangle + \beta^2\langle 2+|J|2+\rangle + 2\alpha\beta\langle 3+|J|2+\rangle \quad (6)$$

The last term can be negative that is, in the coherent mixed valent state interference may occur thereby reducing the effective magnetic moment. For temperatures above 100 K the full moment is obtained in the Curie-Weiss susceptibility; this implies that the $|2+\rangle$ and $|3+\rangle$ states contribute incoherently. A chageover from this coherent to incoherent mixture might also show up as changes in the spin fluctuation spectrum. Experiments to probe these associated dynamics would be most interesting. We should note that the evolution from an incoherent to a coherent mixed valent state corresponds to the crossover from the classical to the quantum regimes in the Fermi liquid picture.

At an early stage, we considered as a possible explanation for the low moment that the orbital moment is effectively quenched so that the magnetism arises only from the spin part of the total moment. For this L=0 case the calculated moment is 1.8 μ_B which is in very good agreement with the observed value. However, the energetics of such an orbital quenching would be difficult to understand since the spin-orbit coupling is larger than the anticipated 4f band-width. In addition, spin-only magnetism should show up as a difference in the form factor as shown by the dotted line in Fig. 3b where the form factor is calculated for L=0[15]. The data, however, agree extremely well with the atomic form factor for pure Tm^{3+}, J=6[15], that is, introducing the orbital moment. This also raises the important problem of why the form factors of Ce and Tm in the mixed valence state correspond so closely to the simple atomic form factors of Ce^{3+} and Tm^{3+}. One expects from simple ideas that the valence mixing would manifest itself in the form factor. As shown in the following paper of Moon et al.[16] the Sm form factor in the mixed valence phase of SmS(Sm^{3+}) is identical to that of the integral valence phase of SmS(Sm^{2+}). It is also not clear why the Sm^{3+} is so modified.

A most interesting result is obtained if we now look at TmSe-2 which, from its lattice parameter should be almost 100% Tm^{3+} and, as discussed above may be thulium deficient[12]. A low temperature diffraction study of the magnetic scattering revealed a completely different magnetic structure from TmSe-1. The new superlattice peaks appear at the (h/2,k/2,ℓ/2) with h, k, and ℓ odd which corresponds to a type II ordering. In this structure, the spins are parallel within the (111) planes but adjacent (111) planes have spins antiparallel (Fig. 5). The structure requires that $|J_{nn}| < 2|J_{nnn}|$ with both exchange integrals antiferromagnetic.

The temperature dependence of these superlattice peaks also show an anomalous behavior as seen in Fig. 5. The intensity is almost linear with temperature which would imply a classical exponent and very different from most other magnetic systems. T_c is also seen to be $\sim$ 4.7 K. Comparing with the weak nuclear peaks, the magnetic moment would be only $\sim$ 0.5 μ_B. Fig. 6 shows the Q scans through the Bragg peak at several temperatures. At the lowest temperature the full width at half maximum (FWHM) is still larger than the resolution. Fig. 7 shows the temperature dependence of the linewidth. The remarkable feature is that it is still almost twice the resolution even at the lowest temperature and is temperature independent for $T < T_N$. From this width we estimate that the correlation length, ξ, does not exceed $\xi = 1/\Delta Q \gtrsim 100$ Å. Another observation is that the critical scattering is observed for a large temperature range above T_c.

This phase transition has indeed very interesting properties. From measurements on similar samples of this material it is still seen to be fcc and from electron microscopy studies it appears to be single phase with no indication of any higher Tm chalcogenides contained in these samples[12]. Measurements by Batlogg et al.[12] show that resistivity and thermal expansion anomalies in this sample are greatly reduced compared to the sample discussed earlier. It is interesting to speculate on the origin of the finite correlation length in this material. Can it be due to defects arising from the Tm deficiency which prevents the long range ordering? It may also be that the delicate balance between the crystal field and exchange parameters exist which prevent the long range ordering from ever becoming complete. Further experiments are planned to study the type of ordering as a function of x, in Tm_xSe.

Since this TmSe sample no. 2 was relatively large ($\sim$ 1 cm^3) inelastic scattering measurements could be performed to probe the spin dynamics and/or crystal field effects. Fig. 8 shows the results at T = 6.0 K for $\vec{Q}$ = (0.65, 0.65, 0.65). There is true inelastic scattering as evidenced by the asymmetric shape of the spectrum as required by detailed balance. The scattering is Q-independent and shows only a slight temperature dependence of the intensity up to 77 K. The spectrum bears a striking resemblance to that observed in γ-$Ce_{1-x}Th_x$[4] only the energy scale is reduced by a factor of $\sim$ 6. This scattering in near integral-valent TmSe most likely reflects the closeness of the f level to the Fermi level and is indicative of the spin fluctuations in the mixed valence systems. The broader

question of the concept of crystal fields[14] in a fluctuating valence systsm is also raised.

Finally, we should comment on the magnetic structures themselves. In sample 1 which has $\sim$ 20%$|2+\rangle$ character, J_{nn} is weak and antiferromagnetic and J_{nnn} is stronger and ferromagnetic. In sample 2 which is assumedly mostly pure $|3+\rangle$, both J_{nn} and J_{nnn} are antiferromagnetic with $|J_{nn}|<2|J_{nnn}|$. Furthermore, from the pressure measurements of Guertin et al.[9], we know that J_{nn} becomes much more strongly antiferromagnetic with decreasing $|2+\rangle$ admixture. The above suggest that as the mixed valence character increases, that is, as the amount of $|2+\rangle$ component increases, an additive ferromagnetic exchange mechanism appears for both nn and nnn. The logical microscopic mechanism[17] is Zener-de Gennes double-exchange which is known to be dominant in such mixed valent d electron systems as $La_{0.7}Pb_{0.3}MnO_3$.

CONCLUSIONS

It is evident that we now have a fairly complete empirical description of the static and dynamic magnetic properties of both $Ce_{1-x}Th_x$ and TmSe, so that we have indeed illuminated our part of Anderson's "mixed valent elephant." We should emphasize, however, that in contrast to Anderson's "blind men" we do not have the benefit of prior experience, that is, $Ce_{1-x}Th_x$ and TmSe exhibit magnetic fluctuation phenomena which are considerably different from all materials studied previously with neutrons, at least, as far as we are aware.

A number of substantive theoretical issues have been raised by our experiments and we hope that a concerted theoretical attack involving real calculations will now be made. In particular, quantitative estimates of the spin fluctuation energy in γ and α $Ce_{1-x}Th_x$ including the dependence on the lattice constant would be most welcome. We also regard the simple behavior observed for the form factors in each of TmSe, $Ce_{0.76}Th_{0.24}$ and SmS as a major puzzle. The question of crystal fields in the mixed valence regime has not yet been addressed theoretically except in the most superficial fashion. From the results in TmSe in particular, one knows that the effective crystal field may be fundamentally altered in the mixed valent state. Finally, more work on the exchange mechanisms, most notably double-exchange, in rare earth mixed valent materials is needed.

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Fig. 1. Valence of Ce and the fractional occupancy of the 4f' level obtained from the lattice parameter measurements (taken from Ref. 4).

Fig. 2. Temperature dependence of the imaginary part of the susceptibility $\chi''(Q,\omega)$ for $Ce_{0.74}Th_{0.2}$ as derived from the observed inelastic neutron scattering spectrum (taken from Ref. 4).

Fig. 3. a) Form factor of Ce in $Ce_{0.74}Th_{0.26}$ just above T_v. The solid line is the calculated $Ce^{3\pm}$ from Ref. 15.

b) Form factor of Tm in TmSe. The solid line is the calculated value for $Tm^{3\pm}$ from Ref. 15. The dotted line is the calculated form factor assuming L=0.

Fig. 4. a) Magnetic structure of monodomain TmSe for a field along the (010) direction. The tetragonal axis is along the z direction.

b) Magnetic phase diagram of TmSe with the magnetic field applied along the (100) direction.

Fig. 5. a) Magnetic structure of TmSe-2.

b) Temperature dependence of the intensity of the (1/2 1/2 1/2) superlattice reflection.

Fig. 6. Q scans through the (1/2 1/2 1/2) magnetic superlattice peak at several temperatures. The right hand scale refers to the 5.0 K data.

Fig. 7. The temperature dependence of the full width at half maximum (FWHM) in $\frac{2\pi}{d}$ $\mathring{A}^{-1}$ units in TmSe-2. The resolution was determined from scans through nuclear Bragg peaks and via calculations.

Fig. 8. Inelastic scan at Q = (0.65,0.65,0.65) for T = 6.0 K in TmSe-2.

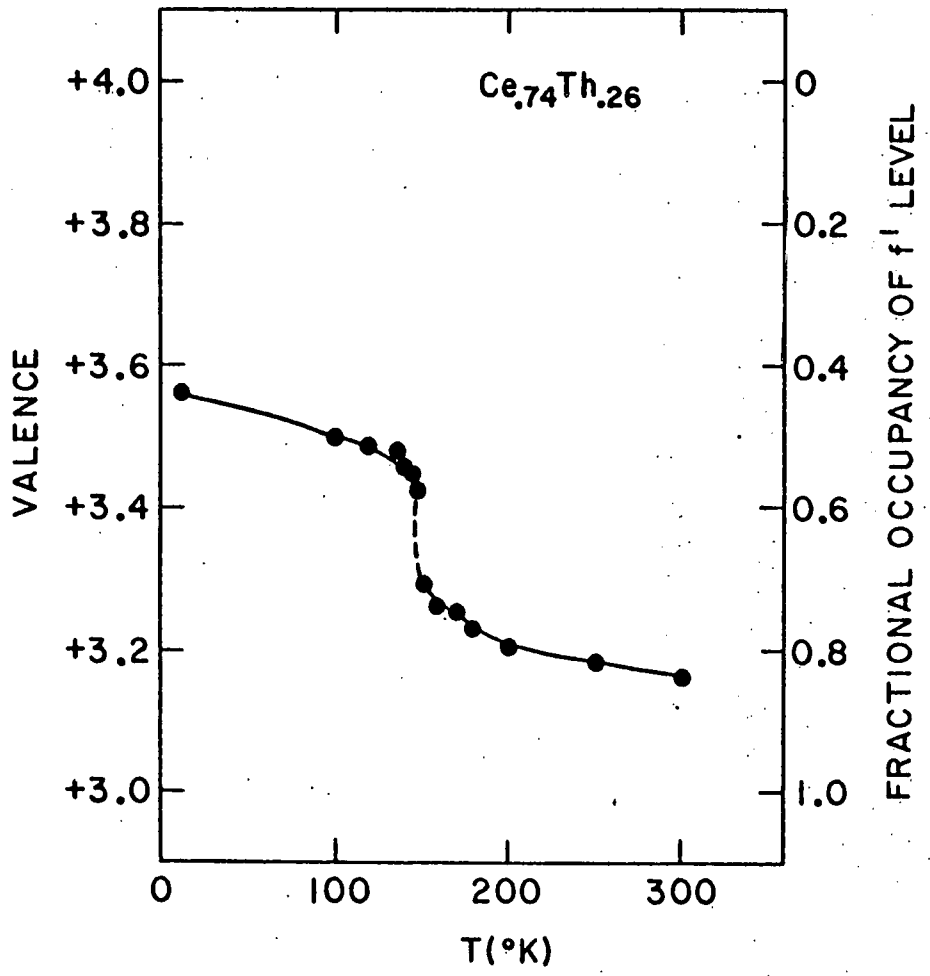

Ce.74Th.26
+4.0
+3.8
+3.6
+3.4
+3.2
+3.0
VALENCE
0
0.2
0.4
0.6
0.8
1.0
FRACTIONAL OCCUPANCY OF f¹ LEVEL
0
100
200
300
T(°K)

FIGURE 1

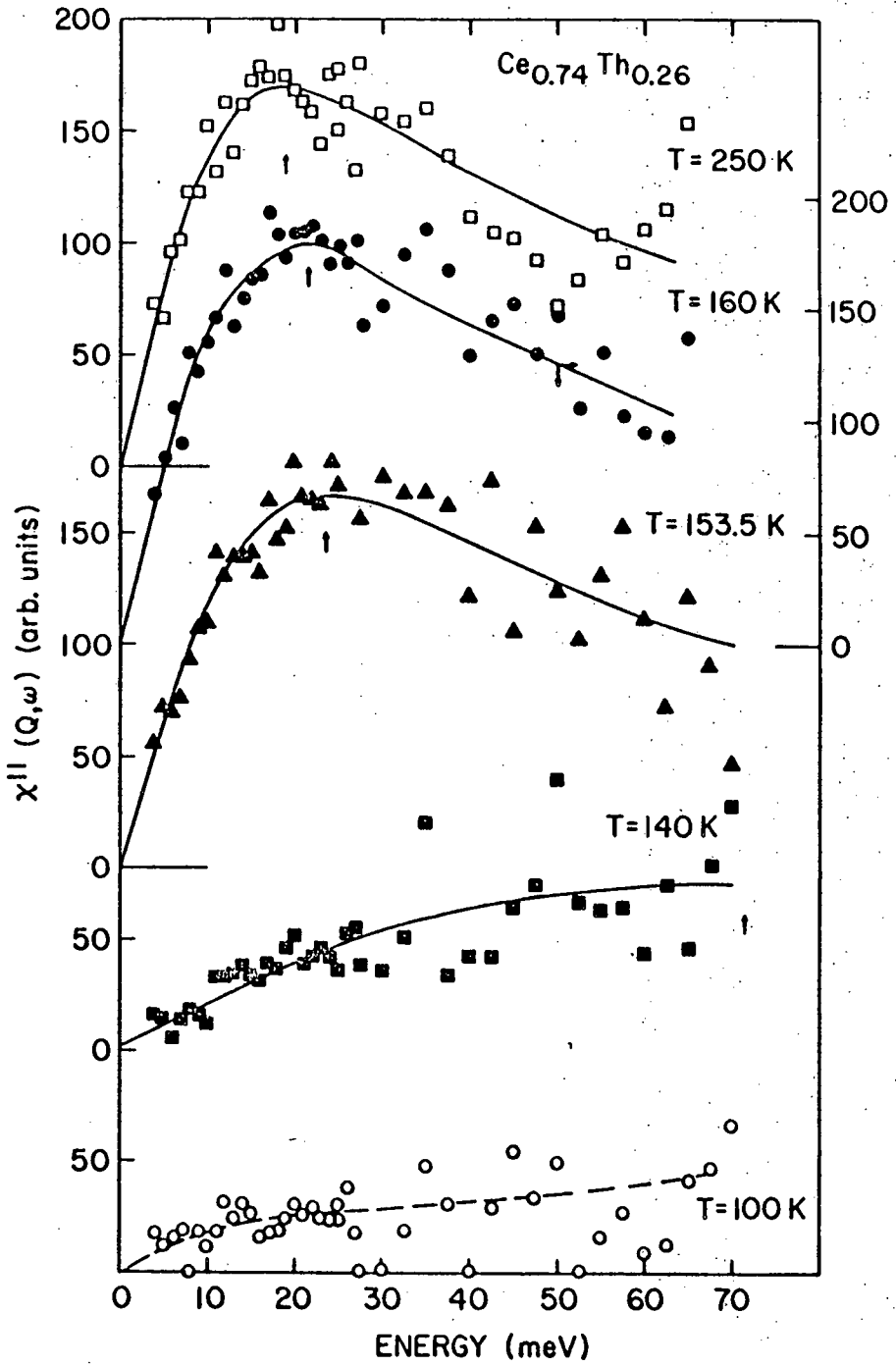

Ce0.74 Th0.26
T = 250 K
T = 160 K
T = 153.5 K
T = 140 K
T = 100 K
χ'' (Q,ω) (arb. units)
ENERGY (meV)
200
150
100
50
0
150
100
50
0
50
0
50
0
10
20
30
40
50
60
70
200
150
100
50
0

FIGURE 2

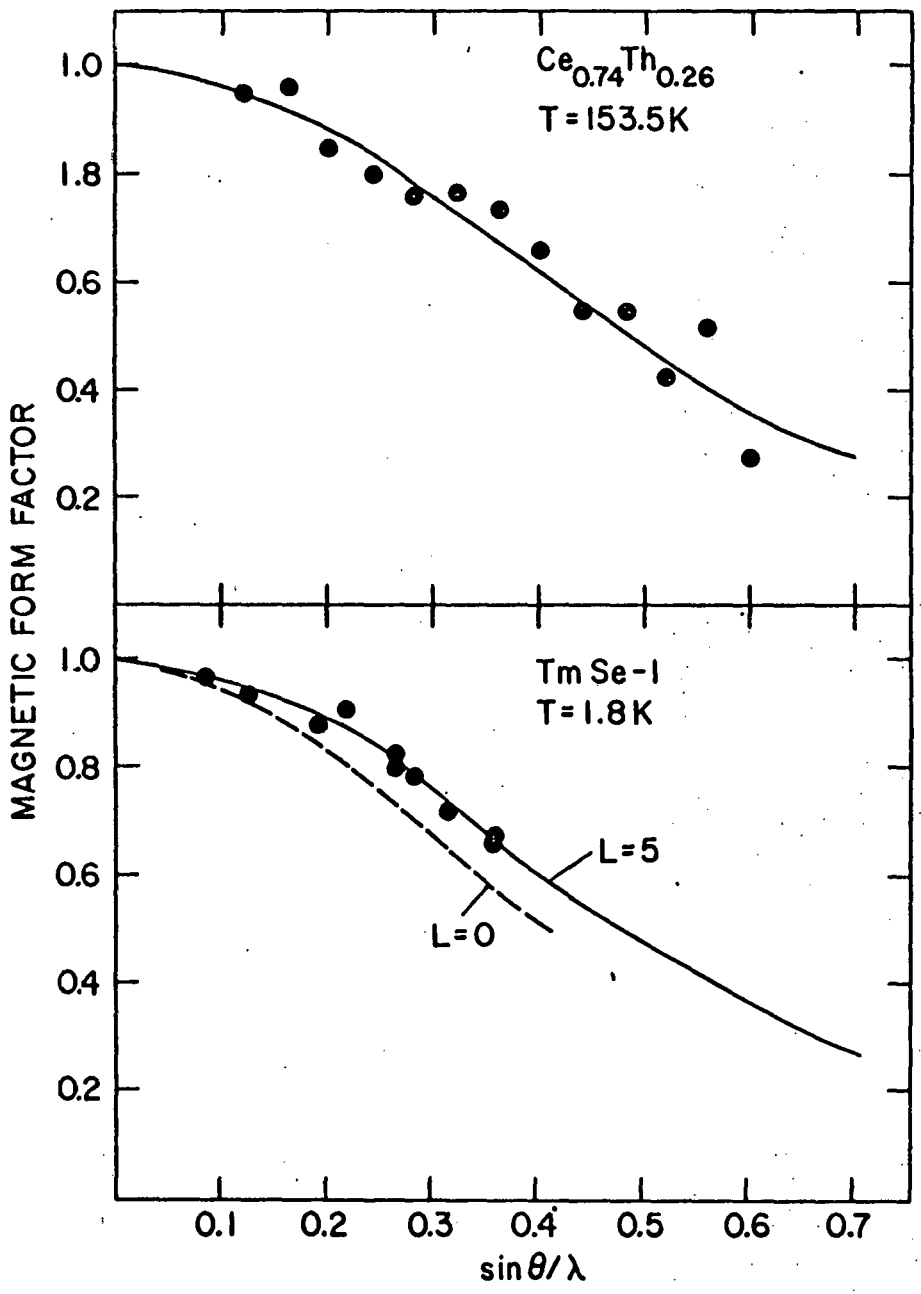
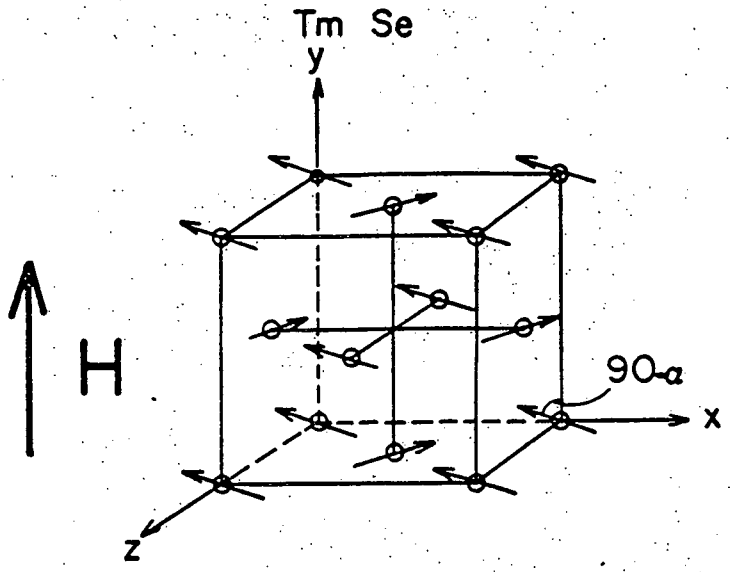

$Ce_{0.74}Th_{0.26}$
T = 153.5K
Tm Se-I
T = 1.8K
L=5
L=0
MAGNETIC FORM FACTOR
sin θ/λ
1.0
1.8
0.6
0.4
0.2
0.8
0.1
0.2
0.3
0.4
0.5
0.6
0.7

FIGURE 3



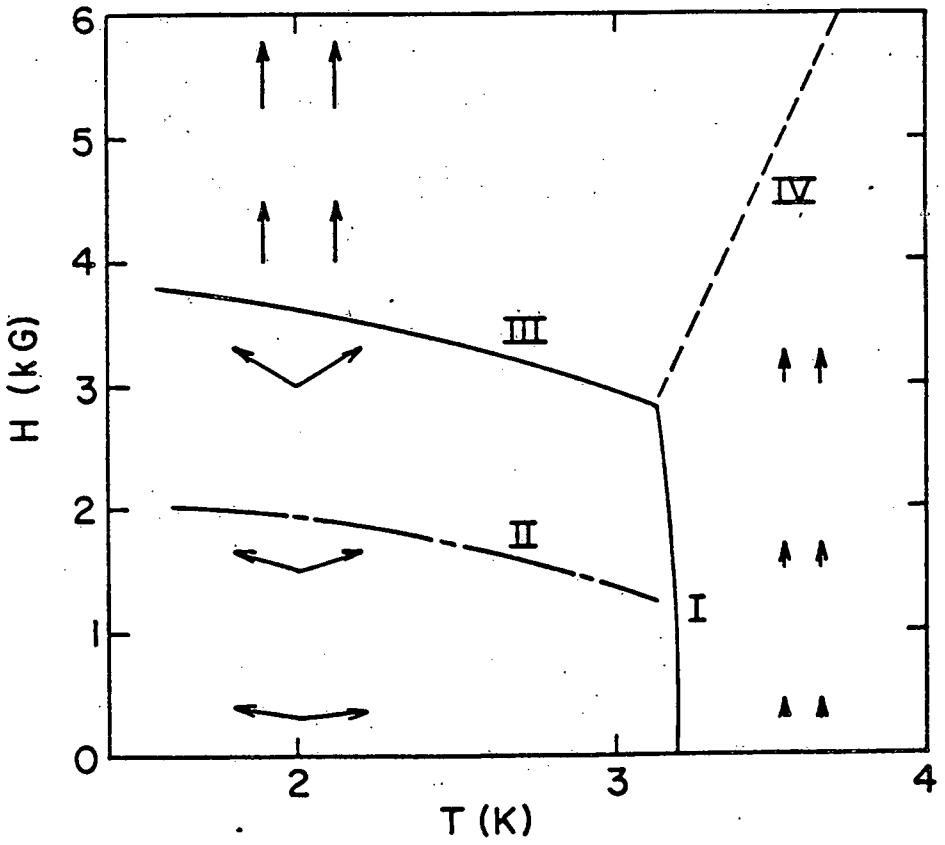


FIGURE 4

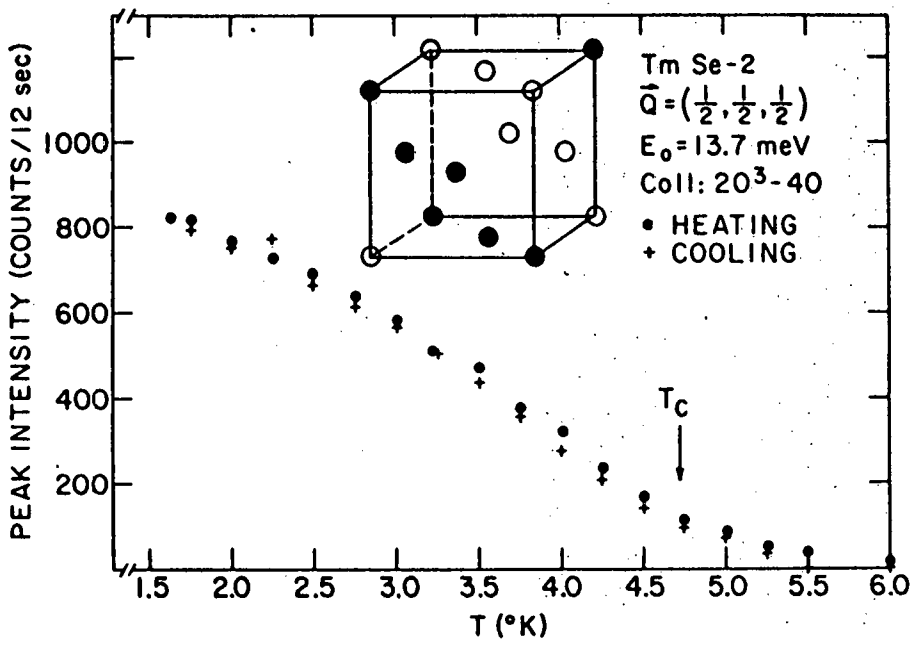

Tm Se-2
$\vec{Q}=(\frac{1}{2},\frac{1}{2},\frac{1}{2})$
E_o=13.7 meV
Coll: 20³-40
• HEATING
+ COOLING
T_C
PEAK INTENSITY (COUNTS/12 sec)
1000
800
600
400
200
1.5
2.0
2.5
3.0
3.5
4.0
4.5
5.0
5.5
6.0
T (°K)

FIGURE 5

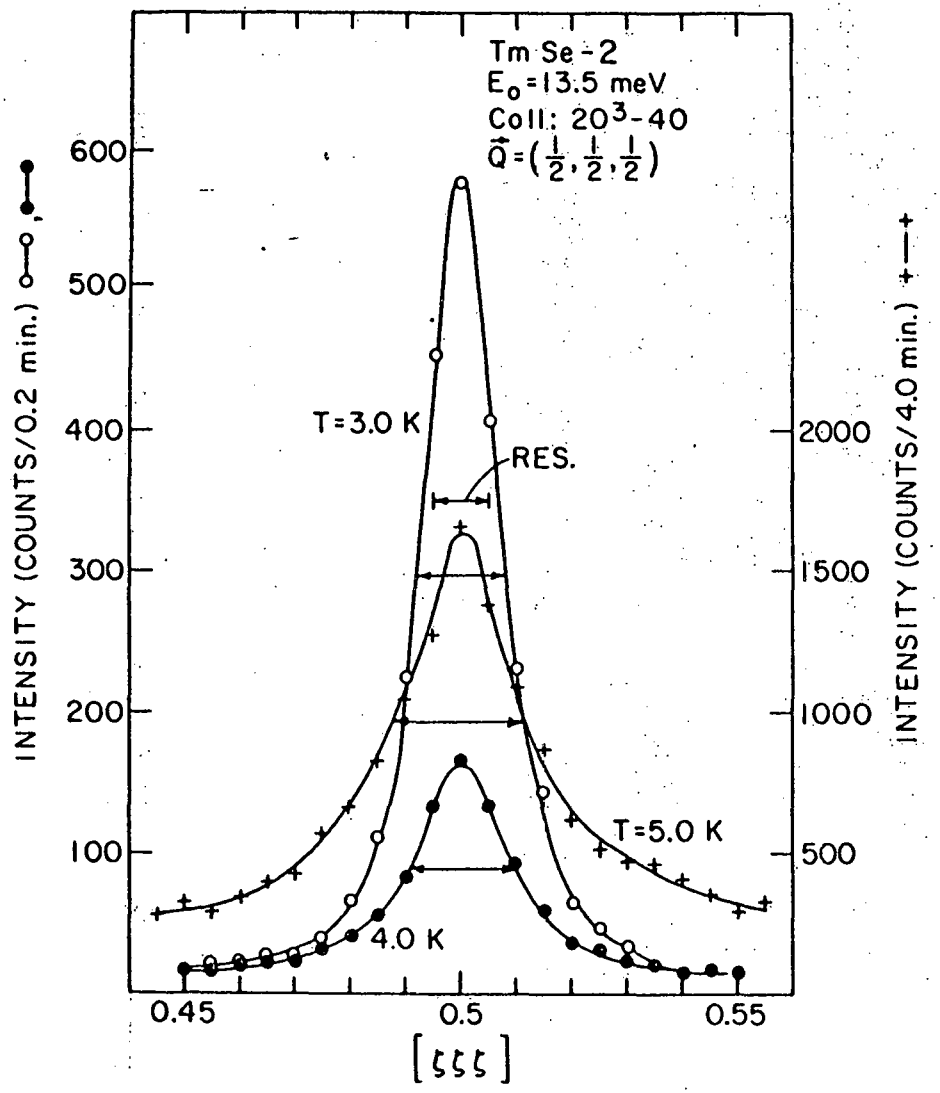

Tm Se-2
E_o=13.5 meV
Coll: 20^3-40
$\vec{Q}=(\frac{1}{2},\frac{1}{2},\frac{1}{2})$
T=3.0 K
RES.
T=5.0 K
4.0 K
INTENSITY (COUNTS/0.2 min.)
INTENSITY (COUNTS/4.0 min.)
600
500
400
300
200
100
2000
1500
1000
500
0.45
0.5
0.55
[ζζζ]

FIGURE 6

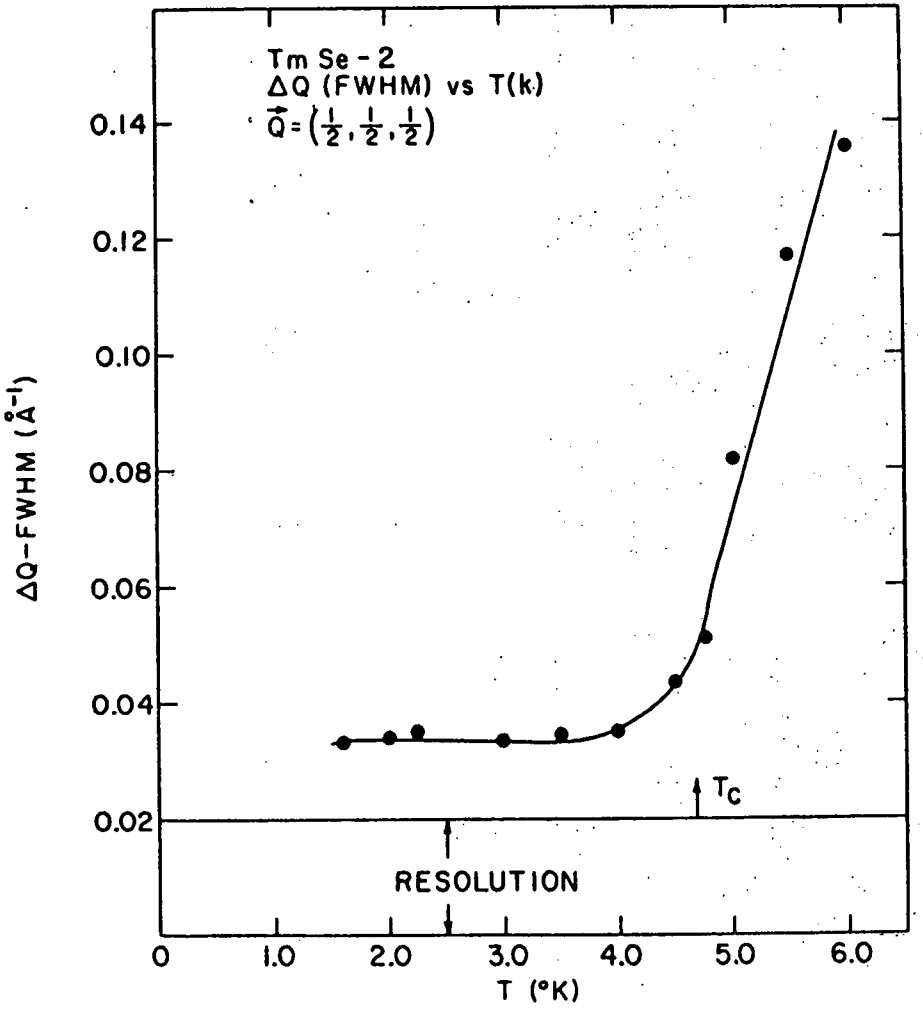

Tm Se - 2
ΔQ (FWHM) vs T(k)
$\vec{Q} = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$
ΔQ-FWHM (Å⁻¹)
0.14
0.12
0.10
0.08
0.06
0.04
0.02
T_C
RESOLUTION
0
1.0
2.0
3.0
4.0
5.0
6.0
T (°K)

FIGURE 7

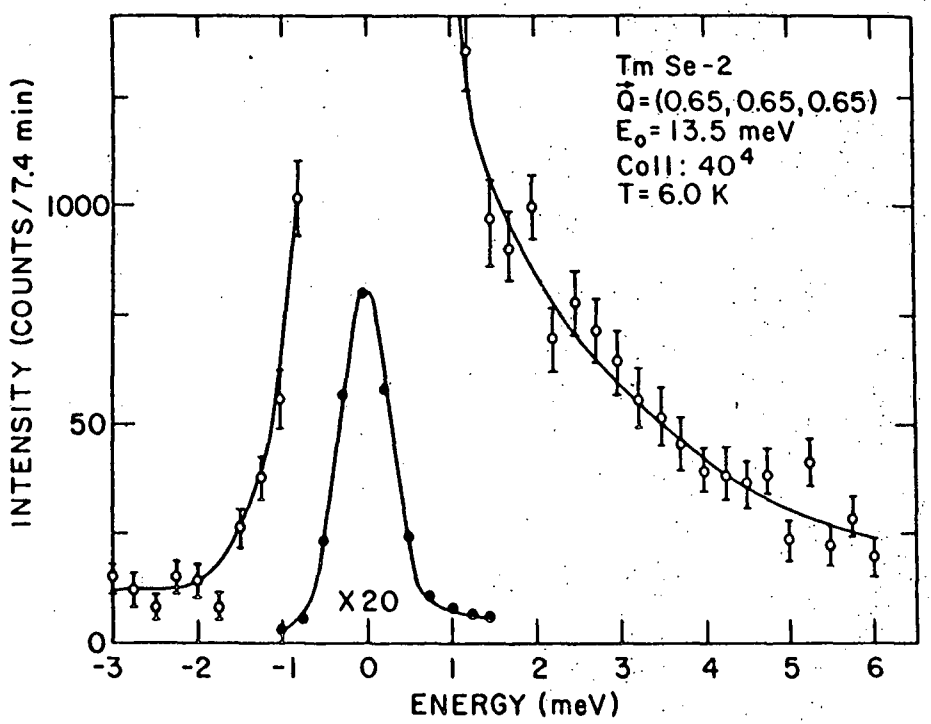

Tm Se-2
$\vec{Q}$=(0.65, 0.65, 0.65)
E_o= 13.5 meV
Coll: 40^4
T= 6.0 K
X20
INTENSITY (COUNTS/7.4 min)
ENERGY (meV)
0
50
1000
-3
-2
-1
0
1
2
3
4
5
6

FIGURE 8