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# Critical Magnetic Scattering from the Heisenberg Ferromagnet EuS

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Abstract

The paramagnetic scattering from the insulating, isotropic ferromagnet EuS is investigated at $T_c$ along the [111] direction by means of inelastic neutron scattering. The energy width of the quasielastic scattering is proportional to $q^z$ with $z = 2.54 \pm 0.10$, in good agreement with the predictions of dynamical scaling theory ($z = 2.5$). z is, however, significantly larger than the value deduced from measurements along the [100] direction ($z \sim 2.2$). Near the zone boundary the magnetic scattering exhibits shoulders the shapes of which deviate from theoretical predictions based on the Heisenberg model.

## I. Introduction

The insulating compounds EuS and EuO have been studied extensively in the past[1] because these systems are almost ideal realizations of the isotropic cubic Heisenberg ferromagnet. The theory of these systems is in a relatively advanced state and many theoretical predictions have been confirmed by experiment[2], for example in EuO, dynamical scaling has been established over four decades in energy[3] and the lineshapes of the magnetic scattering are well reproduced in terms of an effective Heisenberg model by Shastry, Edwards and Young[4,5] near the zone boundary and by renormalization group theory[6] near the zone center[7].

The spin dynamics are expected to be more complicated in EuS than in EuO, because of the competing exchange interactions and the strong dipolar anisotropy[8]. In fact, the spin wave dispersion below $T_c$ is very anisotropic[9]. Recently, Bohn et al measured the q-dependence of the linewidth along the [100] direction at $T_c$ and deduced a dynamical scaling exponent[10] $z = 2.09 \pm 0.06$, in contradiction to the dynamical scaling[11] value $z = 2.5$ (2.46, if Fisher exponent is included). This deviation was attributed to dipolar interactions[10].

In this paper we report on an inelastic neutron scattering study of the spin dynamics of EuS at $T = T_c$, performed in the [111] direction, in order to investigate the anisotropy of the paramagnetic scattering at $T_c$. The new results are compared with recent measurements conducted in the [100] direction[10] and the scattering profiles are compared with some recent theoretical predictions[4,12].

## I. Experimental

The measurements were performed on the same single crystal sample of isotopically enriched $^{153}$EuS used in previous studies[9,10]. EuS crystallizes in the NaCl structure. The lattice constant at $T_c$ is a = 5.95 Å and the nearest neighbour distance $d^*[111]$ = 1.83 $Å^{-1}$. The experiments were performed at the cold source of the Brookhaven High Flux Beam Reactor. We have determined the Curie temperature $T_c$ = 16.6 K by measuring the temperature dependence of the critical magnetic scattering.

The data was collected in the forward direction with fixed final neutron energies $E_f$ = 2.5, 3 or 4 meV. Pyrolytic graphite crystals set for the (002) reflection were used for the (double) monochromator and the analyzer. A cooled Be filter removed higher order neutrons. The energy resolution was between 0.040 meV and 0.14 meV full-width at half-maximum. For more details see Ref. 3.

III. Magnetic Scattering at $T_c$

All measurements of the paramagnetic scattering at $T_c$ were conducted in the constant-Q mode of operation. Some typical scans are shown in Figs. 1 and 2. The nonmagnetic background has been determined by measurements below $T_c$. In Fig. 1 we have subtracted the elastic peak which is mostly due to incoherent scattering (q independent) and amounts to 22% of the magnetic scattering in (a) and to 55% in (b). In Fig. 2 we have subtracted a room background of about 1.5 counts/min from the data. Near the zone boundary $q_{ZB}$ the intensity of the incoherent scattering is about one order of magnitude larger then the magnetic scattering and cannot be subtracted reliably. The magnetic scattering is centered around E = 0 meV, its intensity decreases roughly like $q^{-2}$ and broadens rapidly with increasing q.

We have parametrized the magnetic scattering by fitting the data with the scattering function

$$S(q,E) = 2kT\chi(0) \frac{\kappa_1^2}{q^2 + \kappa_1^2} F(q,E) \frac{E/kT}{1-\exp(-E/kT)} \qquad (1)$$

where $\kappa_1$ is the inverse correlation length, $\chi(0)$ the static susceptibility and $F(q,E)$ a spectral weight function. We have analyzed the small q data by assuming for $F(q,E)$ a Lorentzian

$$F_L(q,E) = \frac{1}{\pi} \frac{\Gamma}{E^2 + \Gamma^2} \qquad (2)$$

where the line width $\Gamma = A\, q^{2.5}$. The solid lines in Fig. 1. represent fits to Eq. (1) with $F(q,E) = F_L(q,E)$, convoluted with the resolution function. The free parameters were a normalization constant, the spin diffusion constant A and the room background.

The data at larger q has been fitted to the three pole approximation[13]

$$F_S(q,E) = \frac{1}{\pi} \frac{\tau\delta_1\delta_2}{E^2\tau^2(E^2-\delta_1-\delta_2)^2 + (E^2 - \delta_1)^2} \qquad (3)$$

with $\tau = \left(\frac{2}{\pi\delta_2}\right)^{1/2}$ and the frequency moments $\delta_1 = \langle E^2\rangle_q$ and $\delta_1\delta_2 = \langle(E^2-\langle E^2\rangle_q)^2\rangle_q^2$. These parameters are known[5]. We compare the profiles predicted by theory[14] with the measurements in Fig. 2. The only free parameter during the fitting procedure was a normalization constant for the magnetic scattering. Both, calculation and data show that shoulders develop at finite energy near the zone boundary. The agreement with theory, however, is much less satisfactory than it was for EuO (Ref.3) and for EuS (q along [100])[10], where theory agrees well with experiment. According to correlation theory[12] (at $T = 1.1T_c$), one would expect a peak at finite energy near $\zeta = 0.3$, which should vanish again at the zone boundary ($\zeta = 0.5$). Our data do not support this either. Both types of theories, based on the Heisenberg model, fail to predict the correct scattering cross sections. The lineshapes will be discussed in another publication[15] in more detail.

In Fig. 3 we have plotted the linewidths versus q in a log-log representation. The linewidths for $\zeta \geq 0.25$ ($q = 0.46$ Å$^{-1}$) have been obtained by fitting the data to Eq.(1) with $F(q,E) = F_S(q,E)$ and determined the half-width at half-maximum (HWHM) from the fitted parameters $\delta_1$ and $\delta_2$ as described in Ref. 3. Above $\zeta = 0.30$ ($q = 0.55$ Å$^{-1}$) the HWHM starts to saturate. Therefore we have determined the critical exponent z by fitting the line widths to the power law $\Gamma = A\, q^z$ taking into account the data for $0.09$ Å$^{-1} \leq q \leq 0.46$ Å$^{-1}$ only. We obtained $A = 2.1 \pm 0.3$ meV $A^z$, $z = 2.54 \pm 0.10$, in good agreement with dynamical scaling theory[11] which predicts $z = 2.5$.

## IV. Discussion

Neutron measurement along the [100] direction in EuS at $T_c$ have recently been extended to smaller q by means of neutron spin echo techniques. The line widths are proportional to $q^{2.2}$ over nearly four decades in energy[16]. We have indicated these results in Fig. 3 by means of a dotted line. The line widths for $q \leq 0.2$ Å$^{-1}$ are isotropic, where as for $q \geq 0.25$ Å$^{-1}$ they are larger along [111] than along [100]. This apparent discrepancy can be traced back to the use of a double Lorentzian spectral weight function[17] in Ref. 10. In fact, the magnetic scattering for q along [100] extends to higher energies than for q along [111] as expected, since the spin wave energies below $T_c$ are also larger[9] along [100].

The most important result from the preceeding section is the good agreement of the exponent z with dynamical scaling theory for q along [111]. The exponent is, however, significantly larger than $z = 2.2$ (Ref. 16) or $z = 2.09 \pm 0.06$ (Ref. 10), the measured values deduced from line width measurements with q along [100]. The latter value has been explained by dipolar interactions. Dipolar dynamics are expected to dominate the isotropic critical behavior ($z = 2.5$) for $q < q_D = 0.27$ Å$^{-1}$ and z is expected to cross over to 2 (Ref. 18). The dipolar wave vector $q_D$ has recently been verified directly in EuS and EuO by polarized neutron scattering[19].

One may speculate that there should be no dipolar crossover at all at $T_c$ along [111] by the following reason: The spin wave specta at low temperatures exhibit no energy gap along [111], because [111] is the easy axis of magnetization. Therefore there is no dipolar contribution to the exchange energy. On the other hand, our new data may also be interpreted in the following way. Above $q = q_D$ the system is in the isotropic region where dynamical scaling is valid ($z = 2.5$). For $q < q_D$ we observe the dipolar cross over to $z = 2$. In order to test the above conjectures it is necessary to extend the measurements to smaller q along [111].

We do not understand, why no dipolar crossover has been observed in neutron scattering studies in EuO, since $q_D = 0.15$ Å$^{-1}$ lies well within the q range investigated. Is it the competing exchange interactions in EuS, which are absent in EuO ? We hope that the present line width measurements will be extended to smaller q along different symmetry directions in EuS and EuO in order to obtain a better understanding of the influence of dipolar interactions on the critical dynamics.

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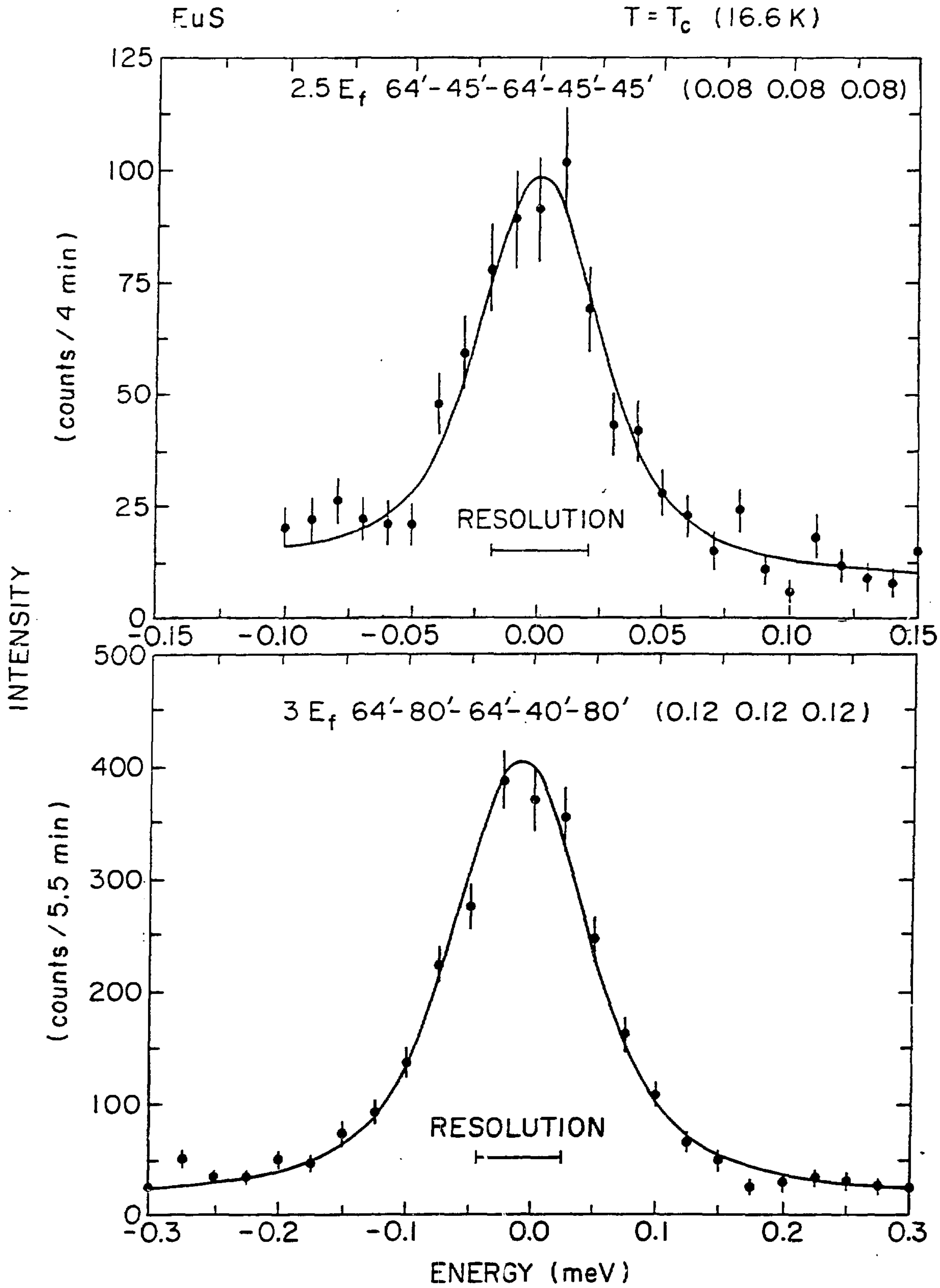
## Figure Captions

1. Typical constant-q scans performed at $T_c$. The elastic background has already been subtracted. The solid lines are fits to the cross section (Eq.(1)), assuming a Lorentzian spectral weight function.
2. Constant-q scans performed near the zone boundary. The room background has already been subtracted. The shoulders at finite energy are more ponounced than the theory predicts (solid lines).
3. The data points represent line width measurements along the [111] direction. The broken line indicates measurements along the [100] direction[10,16] with $z = 2.2$. The arrow at $q_D = 0.27$ Å$^{-1}$ indicates the dipolar wave vector. The horizontal bars indicate the energy resolution of the various experimental setups.



EuS
T = Tc (16.6 K)
2.5 Ef 64′-45′-64′-45′-45′ (0.08 0.08 0.08)
RESOLUTION
3 Ef 64′-80′-64′-40′-80′ (0.12 0.12 0.12)
RESOLUTION
INTENSITY
(counts / 4 min)
(counts / 5.5 min)
ENERGY (meV)
125
100
75
50
25
0
500
400
300
200
100
-0.15
-0.10
-0.05
0.00
0.05
0.10
0.15
-0.3
-0.2
-0.1
0.0
0.1
0.2
0.3


**Figure 1**

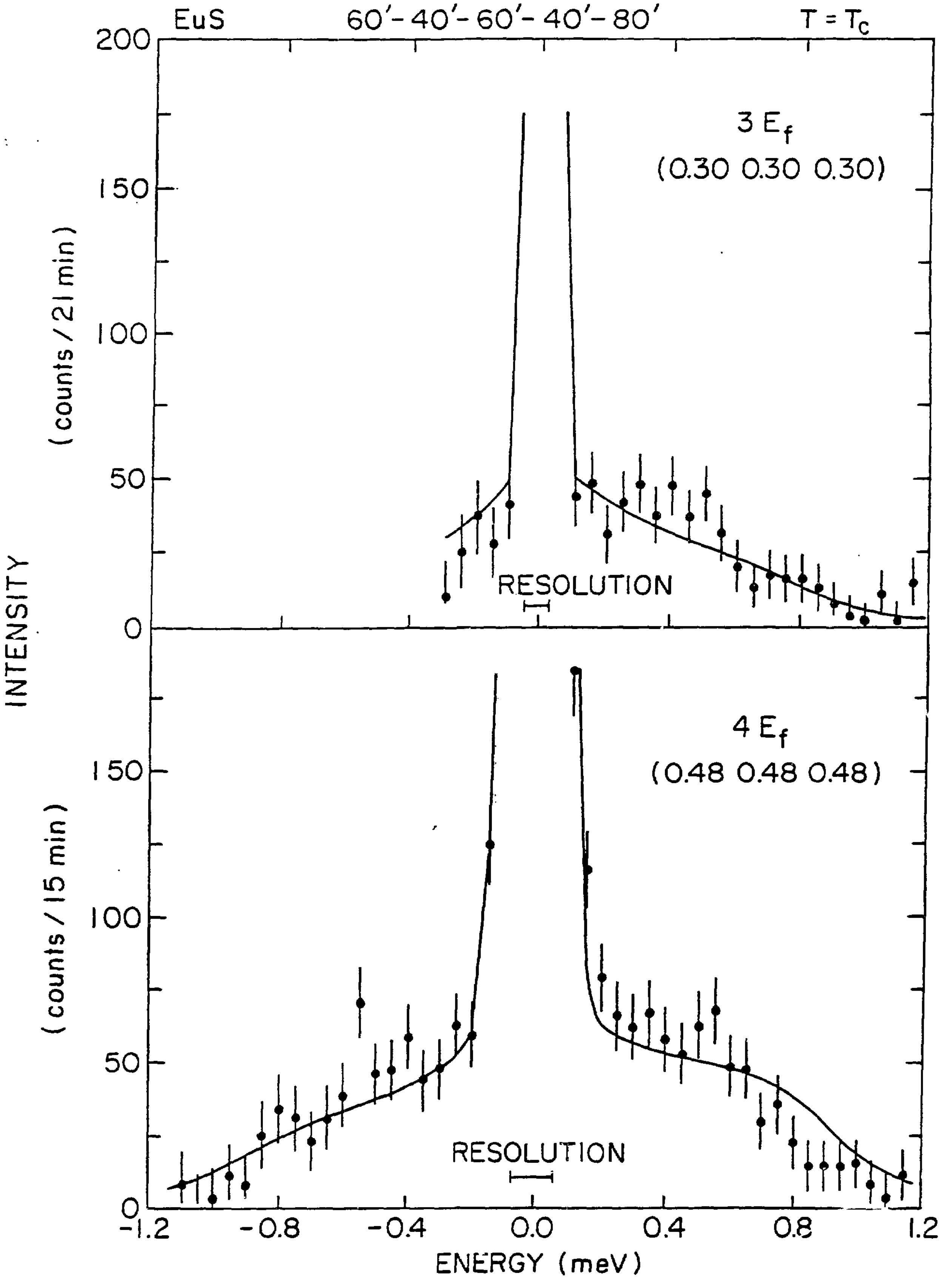

EuS
60′-40′-60′-40′-80′
T = Tc
3 Ef
(0.30 0.30 0.30)
RESOLUTION
4 Ef
(0.48 0.48 0.48)
RESOLUTION
INTENSITY
(counts / 21 min)
(counts / 15 min)
ENERGY (meV)
200
150
100
50
0
-1.2
-0.8
-0.4
0.0
0.4
0.8
1.2

**Figure 2**

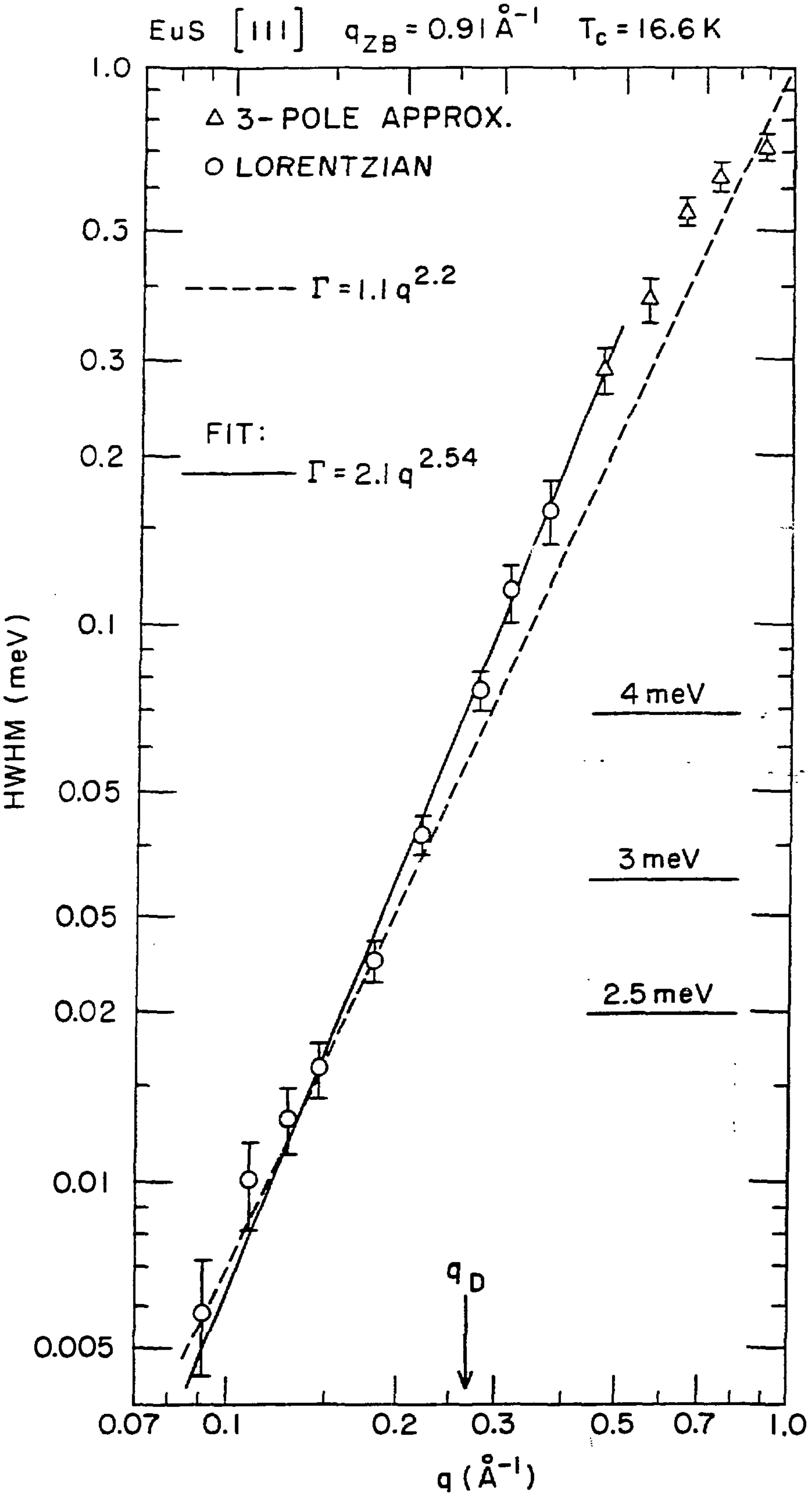

EuS [111] $q_{ZB}$ = 0.91 Å$^{-1}$ $T_c$ = 16.6 K
△ 3-POLE APPROX.
○ LORENTZIAN
$\Gamma = 1.1 q^{2.2}$
FIT:
$\Gamma = 2.1 q^{2.54}$
4 meV
3 meV
2.5 meV
$q_D$
HWHM (meV)
q (Å$^{-1}$)
1.0
0.5
0.3
0.2
0.1
0.05
0.05
0.02
0.01
0.005
0.07
0.1
0.2
0.3
0.5
0.7
1.0

**Figure 3**