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THE CENTRAL PEAK REVISITED

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Abstract

The central peak in SrTiO_3 was first observed by Riste and his collaborators in 1971. This was one of the key discoveries leading to an understanding of the dynamics of phase transitions. The most recent discovery of two length scales in SrTiO_3 motivated a reinvestigation of the soft phonon and associated central peak by neutron scattering. These recent experiments shed new light on the nature of the central peak. It is now well established to be strongly sample dependent and it originates from defects in bulk crystals.

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I. INTRODUCTION

Emil Samuelsen gave a very interesting historical background of the discovery of the central peak by Riste et al⁽¹⁾ in 1971. Looking back, the report was a real surprise for me at that time. In 1969, neutron scattering studies^(2,3) established that the zone boundary soft mode was the driving mechanism of the 100K transition. The soft phonon at the R point, as shown in Fig. 1, is strongly temperature dependent and the energy decreases toward zero as the transition temperature is approached from higher temperatures. The soft mode picture of the transition looked simple, elegant, and complete.

Soon after, in 1971, Riste et al. reported that there is an additional important element present near the phase transition. The so-called central peak, which was not observed in the earlier works, appears⁽¹²⁾ as a very sharp peak, at the zero energy transfer. This sharp component diverges as T_c is approached while the side bands with mode frequency ω_∞ saturate at a finite energy. Why did we miss such an important cross section in the original neutron measurements? This central peak appears at the elastic R point, where the observed intensities are contaminated by higher orders. It took Riste's intuition and understanding of the instrument to dig in and discover the central peak.

A detailed characterization was immediately carried out by Shapiro et al.⁽⁴⁾, in collaboration with Riste, in 1972. Typical examples of the spectra are shown in Fig. 1. The case for KMnF_3 demonstrates the example of overdamped soft mode. Even in this case, the central peak is clearly seen. Similar central peaks have subsequently been observed in many different types of phase transition. A particularly striking example⁽⁵⁾ is the case of Nb_3Sn (Fig. 2), where the transverse acoustic modes soften as the temperature approaches the cubic-tetragonal transition temperature of 45K.

The strength of the central peak can be characterized by a coupling constant δ^2 as shown in Fig. 3. For temperatures not too close to T_c , the total integrated intensity can be divided into two parts;

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$$I_{TOT} = I_{central} + I_{phonon}$$

$$\frac{1}{\omega_0^2} = \frac{\delta^2}{\omega_0^2 \omega_\infty^2} + \frac{1}{\omega_\infty^2} \quad (1)$$

This yields

$$\delta^2 = \omega_\infty^2 - \omega_0^2 \quad (2)$$

This coupling constant shows some temperature dependence, as shown in Fig. 3(b). Near the phase transition where $\kappa_c = 0.64 \kappa_0$ then $\delta^2 = \omega_\infty^2$.

Despite the extensive studies, the true nature of the central peak has never been well understood. Certain theories state it is dynamical due to anharmonic process⁽⁶⁾, while other state it is related to defects in the sample⁽⁷⁾. The well established elastic nature of the central peaks contradicts the dynamical origin and some measurements⁽⁸⁾ suggested a defect origin. Experimentally, Cowley and Shirane⁽⁹⁾ established that the central peak in SrTiO₃ originates from the bulk of the material, and not the outer surface region or "skin" of the sample.

II. Two Length Scales in SrTiO₃

In 1986, Andrews⁽¹⁰⁾ performed a high resolution x-ray study of SrTiO₃ and reported a new "sharp" peak in SrTiO₃ as shown in Fig. 3(d). This is called the two length scale problem because the profile in Fig. 3(d) reveal two correlation lengths, A short length associated with the broad peak and a long length scale associated with the sharp narrow peak. A further study of these profiles were reported by McMorro et al⁽¹¹⁾. These measurements were carried out by using high-resolution x-ray scattering techniques. It is a sharp peak in q space, in contrast to the sharp central peak in E-space. This sharp new peak appears at a narrow temperature range above the transition temperature.

This sharp peak is superimposed on a broad peak which is expected from the soft phonon dispersion curve. Again, when the transition temperature is approached from above, the intensity of this sharp component diverges. Immediately, the question was asked whether this sharp q peak "is" the central peak itself. When we looked into this question in 1992, we realized then that the central peak had never been characterized properly in q space. All previous neutron experiments probed the E dependence of the central peak. The q-dependence of the central peak is the focus of recent paper by Shirane et al⁽¹³⁾.

III. q Dependence

The identical SrTiO₃ crystal (Sanders), which was used in 1971 experiment⁽⁴⁾, was again reexamined carefully and results are shown in Fig. 4 and 5. Because of the extremely sharp energy width of the central peak (see Fig. 1), it is easy to separate out this component from the soft phonon by judicious selection of neutron energy and spectrometer collimation.

The soft phonon branch has a dispersion as q moves away from the ZB

$$\omega_{\infty}^2(q, T) = \omega_{\infty}^2(0, T) + \alpha_q q^2 \quad (3)$$

Then we can define two inverse correlation length κ_{α} and κ_0 related to the q-widths of the phonon part and the total intensity.

$$\kappa_{\infty}^2 = \frac{\omega_{\infty}^2(0, T)}{\alpha_q} \quad (4)$$

$$\kappa_0^2 = \frac{\omega_0^2(0, T)}{\alpha_q} \quad (5)$$

x-ray scattering measures the total energy integrated scattering and measured values of k_{L1} , (dotted lines in Fig. 5) are in good agreement with the neutron results of κ_0 , which includes the central peak and the phonons. Note that the two different directions [100] and [011] have noticeably different dispersion slopes (Fig. 4) and these are clearly reflected in the anisotropy of the central peak intensities shown in Fig. 4.

Now the q width of the central peak can be expressed as (using the above equations)

$$I_{cent} = \frac{T}{\alpha_q^2} \left[\frac{\delta^2}{(\kappa_1^2 + q^2)(\kappa_-^2 + q^2)} \right] \quad (6)$$

At high temperatures when $\omega_\infty^2 \gg \delta^2$ and consequently $\omega_0^2 = \omega_\infty^2$, the central peak has a Lorentzian-squared line shape

$$I_\infty \frac{\delta^2}{(\kappa_0^2 + q^2)^2} \quad (7)$$

The half width at half maximum of this central peak line shape is

$$\kappa_c = 0.64 \kappa_0 \quad (8)$$

The temperature dependence of κ_c is a unique signature of the central peak and a quantity that can be easily verified experimentally (see Fig. 5).

These results shown in Fig. 4 and 5 demonstrate conclusively that the central peaks are not sharp at all in q space and related to the broad peak observed in x-ray experiments. Thus, its origin is entirely different from that

of the sharp q peak. Meanwhile, a series of experiments⁽¹³⁻¹⁵⁾ demonstrated decisively that the sharp q peaks originate from the outer skin of the crystal, whereas the central peak is a bulk property⁽⁹⁾.

IV Sample Dependence

Our recent measurements of the central peak have clarified the distinction between the central peak and the sharp q peak. They also provided a better characterization of the central peak as the integral part of the phonon spectral weight. Very recently, further studies of the central peak were carried out by Hirota et al⁽¹⁴⁾. They reported systematic x-ray and neutron scattering measurements on several differently prepared single crystals of SrTiO₃. The main focus of this study was to characterize and clarify the origin of the narrow q peaks. However, the central peak was also measured as part of the overall characterization.

The original crystal used in ref. 4 and 8 was a bulk crystal obtained from Sanders Associates. This was grown by the top-seeded method, which yielded an essentially strain-free crystal with an effective mosaic spread of 0.017° determined from x-ray diffraction and 0.004 from γ-ray diffraction⁽¹⁶⁾. The crystal is transparent but has a brownish color of unknown origin. Recently, SrTiO₃ has become widely used as a substrate for high T_c superconducting thin films. High quality samples are now commercially available. These are grown by the Verneuil method which has been greatly improved in recent years. These crystals have narrow mosaic and they are clear without brownish tint.

The relative strength of the central peaks in various crystals was quantitatively characterized by the coupling constant δ^2 which can be determined by the relation⁽¹²⁾

$$\delta^2(q, T) = \frac{R}{R+1} \omega_{\infty}^2(q, T) \quad (9)$$

when $R = I_{\text{central}}/I_{\text{phonon}}$. New crystals show substantially smaller

δ^2 as seen in Fig. 6. Hence, δ^2 is presented as a function of ω_{∞}^2 . It is likely that modern, clear crystals have less defects because of improved growth technique. Thus we can consider Fig. 6 as the additional support for the defect model for the central peak. It is rather amusing to see that the high T_c research has helped to understand the central peak!!

It was in 1957 when I first met Tormod Riste when he visited Brookhaven. Since then, it has been a long and enjoyable association over the past 38 years. I would like to thank Tormod for many stimulating discussions on the central peak and many other topics. I have also benefited by discussions with R. A. Cowley, Y. Fujii, K. Hirota, J. R. Schneider, and S. M. Shapiro. This work was supported in part by the U.S. Japan Collaboration for Neutron Scattering. Work at Brookhaven was carried out under Contract No. DE-AC02-76CH00016 Division of Materials Science, U.S. Department of Energy.

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Figure Captions

- Fig. 1. High resolution neutron scattering cross sections for the soft mode and central peak. After Shapiro et al.⁽⁴⁾.
- Fig. 2. The central peaks in Nb₃Sn near the 45K transition. After Axe and Shirane⁽⁵⁾.
- Fig. 3. Characterization of the central peak (a), (b) (c) and the newly discovered narrow q peak in SrTiO₃. After Shirane et al.⁽¹²⁾.
- Fig. 4. Phonon dispersions and central peaks in SrTiO₃. After Shirane et al.⁽¹²⁾.
- Fig. 5. Three inverse correlation length κ_{∞} , κ_{∞} , κ_c in SrTiO₃. After Shirane et al.⁽¹²⁾.
- Fig. 6. Sample dependence of the coupling constant δ^2 in SrTiO₃. After Hirota et al.⁽¹⁴⁾.

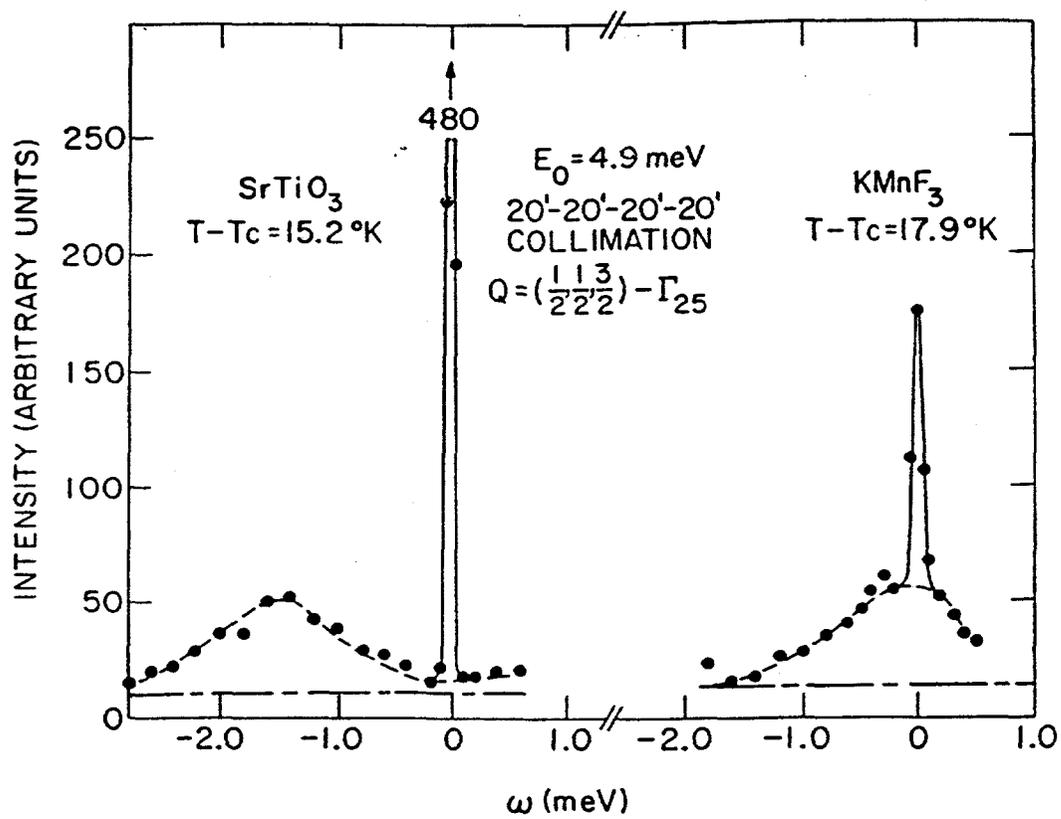


FIGURE 1

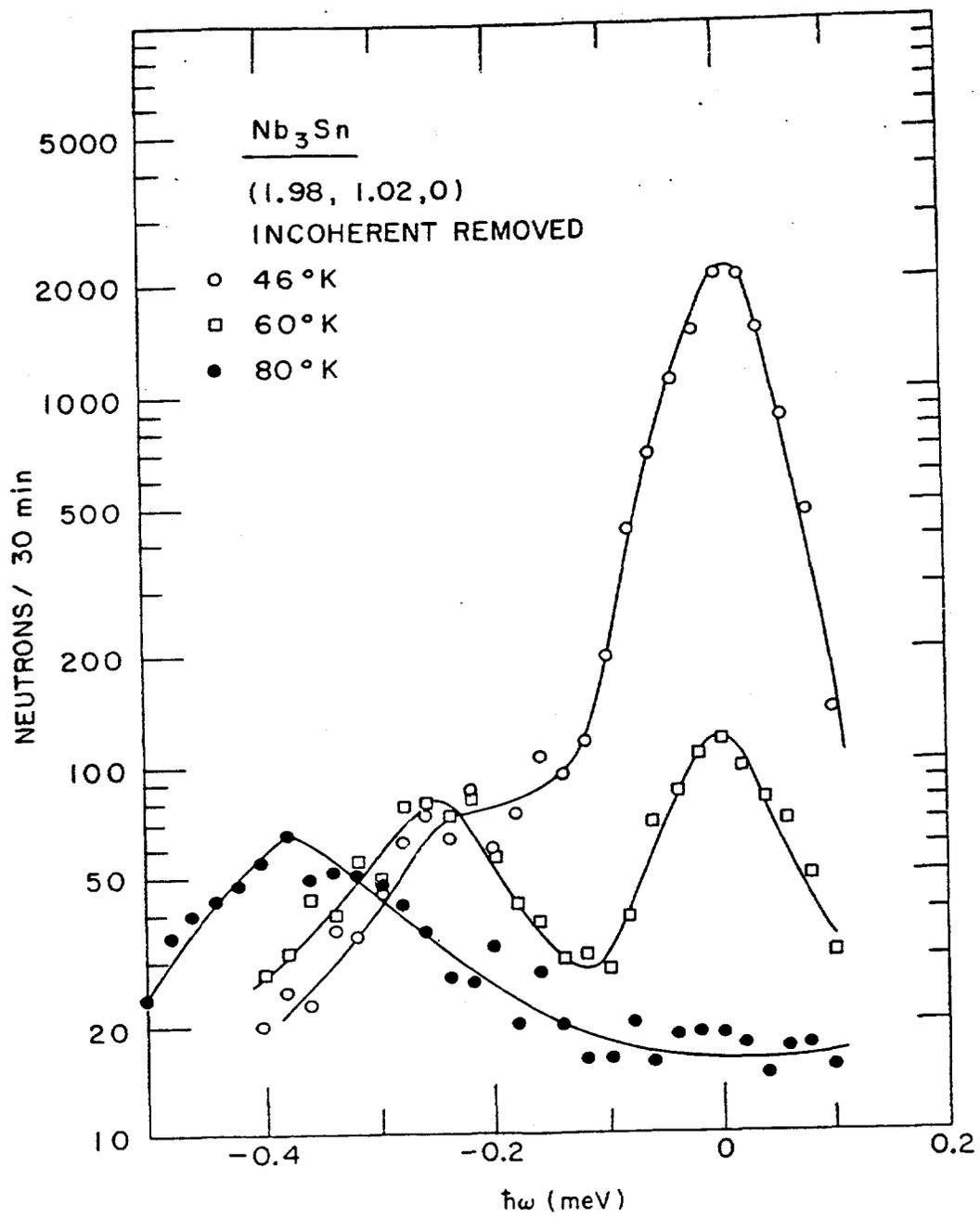


FIGURE 2

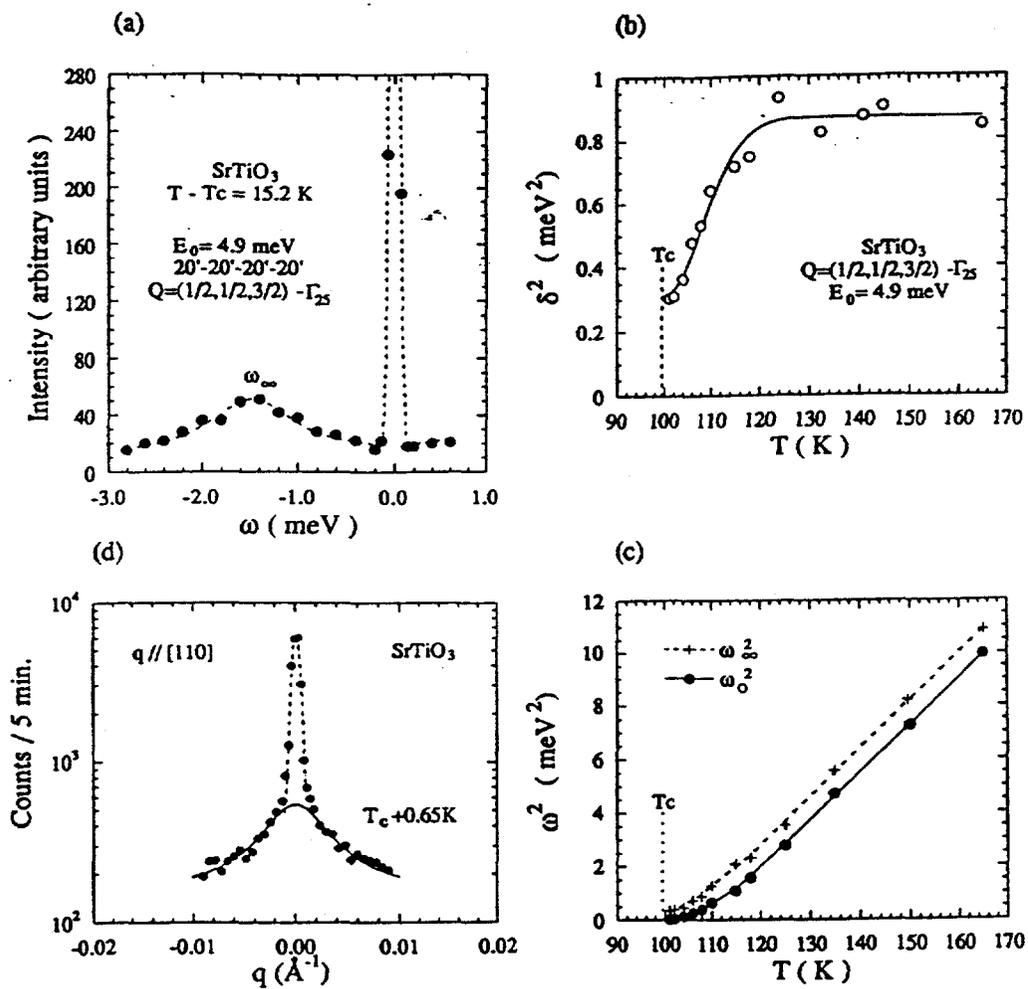


FIGURE 3

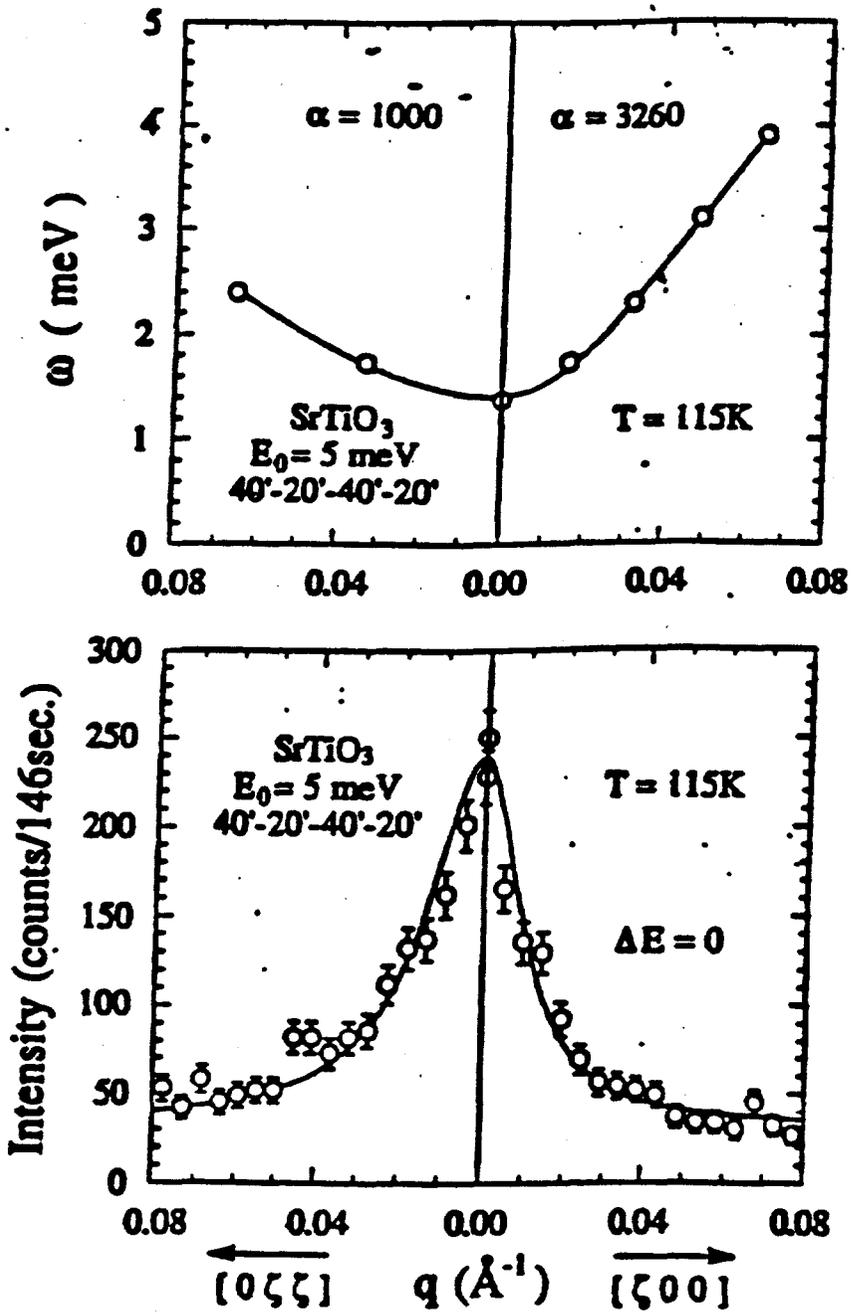


FIGURE 4

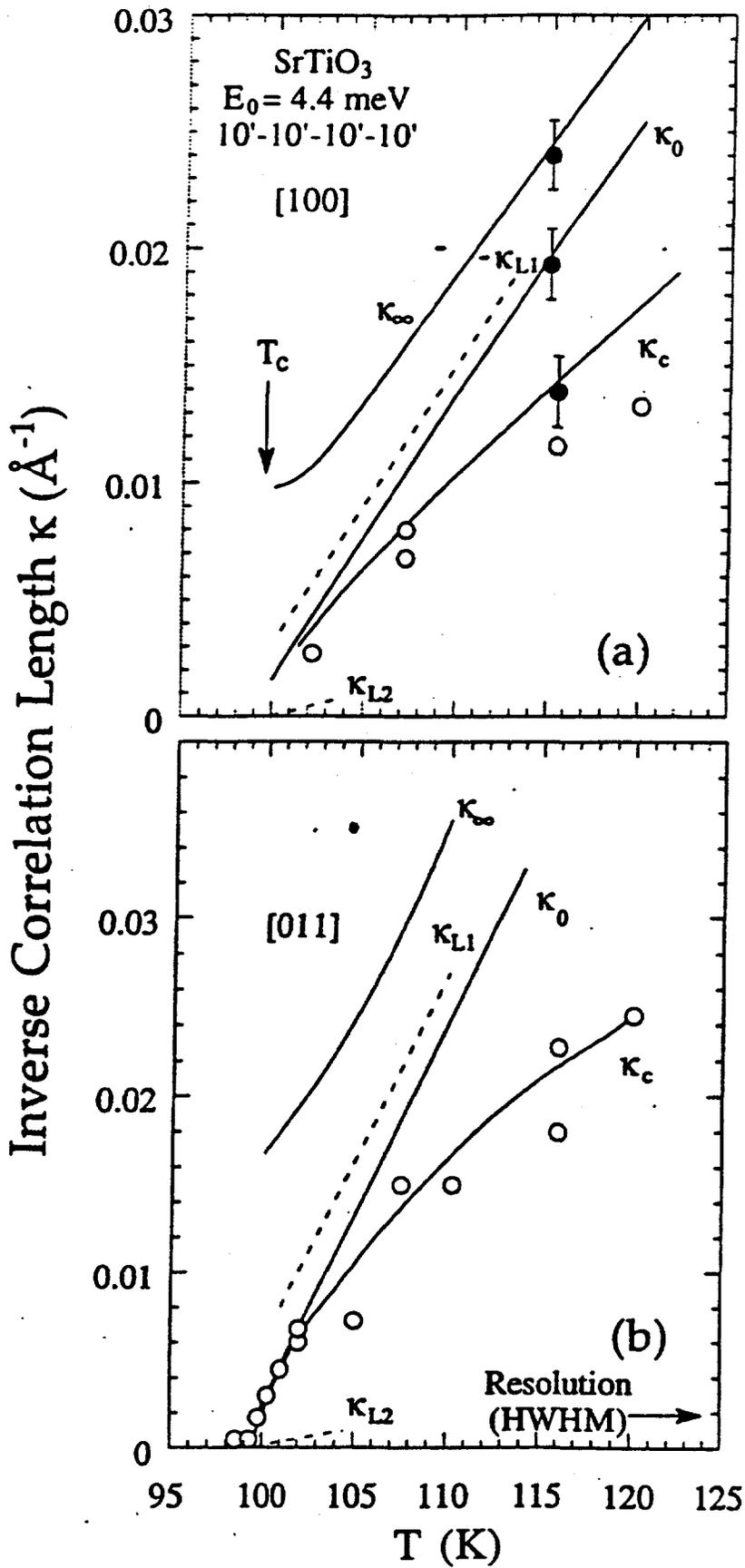


FIGURE 5

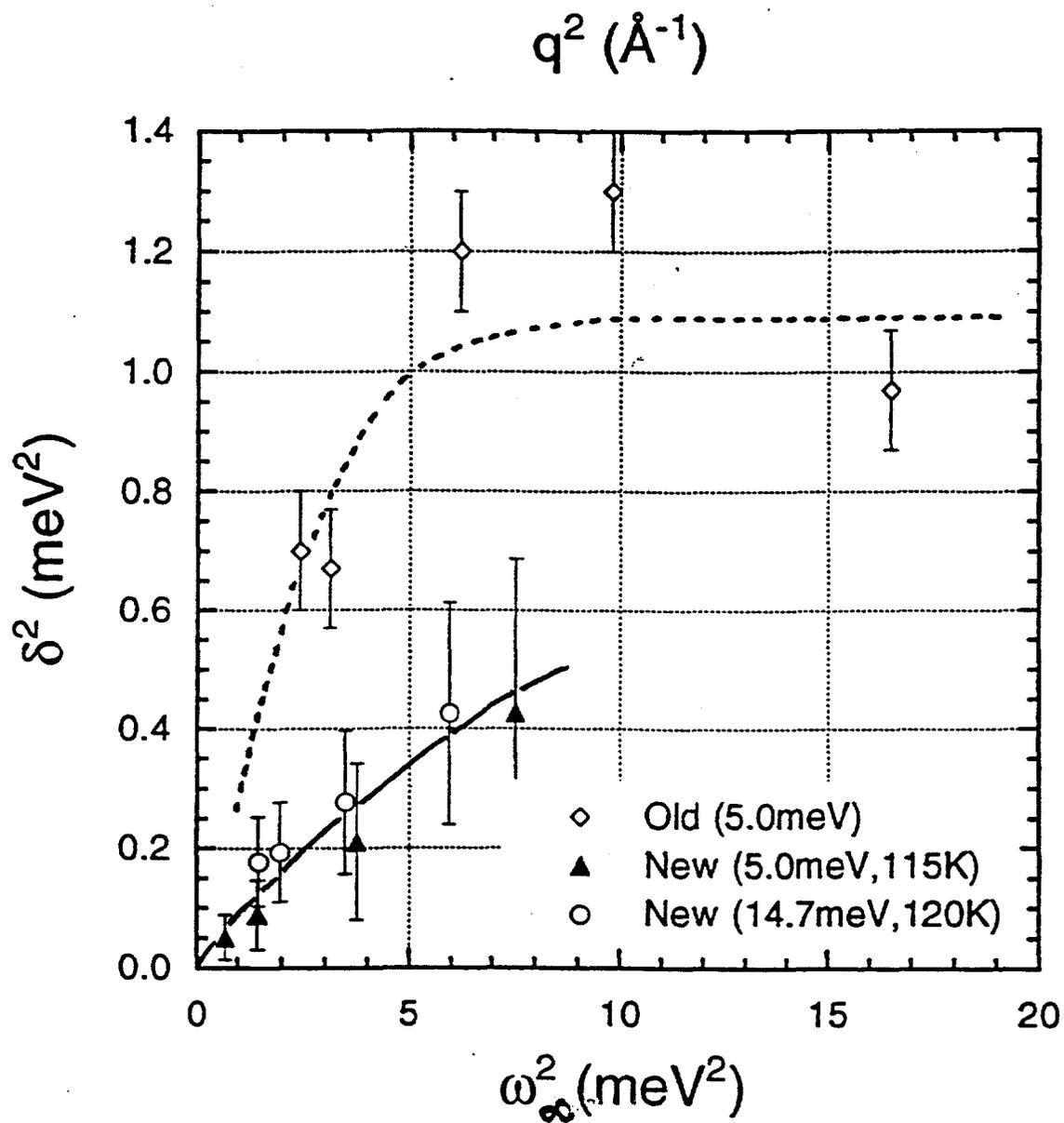


FIGURE 6