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Comparison of Kinematic and Optical Matrix Methods for Calculating Reflectivity Profiles of Solid Thin Films and Multilayers

J Eastoe and J Penfold

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Abstract

A comparison of the calculation of neutron reflectivity profiles R(Q) by optical matrix methods and a multilayer method, based on the kinematic approximation, has been made. The latter method provides a fast simple route to calculating R(Q) and may be used with confidence for systems of total thickness $\lesssim 200$ Å for a scattering length density difference $\Delta\rho$ of the order of 1.0×10^{-5} Å^{-2}. For thicker layers the approximation is poor and the optical matrix method must be used for an accurate evaluation of such reflectivity data over a wide Q range .

1. Introduction

The specular reflection of neutrons is now being extensively applied to a range of problems for the characterisation of thin films and multilayers [1].

To date, the majority of reflectivity data has been evaluated by model fitting [2,3] using calculation based on the optical multilayer methods [4]. This approach provides a particularly convenient method for calculating exactly reflectivity profiles for any given discrete density profile, and where surface and interfacial imperfections can be easily included. However for a large number of layers, as can be often encountered in multilayer samples, the method can consume large amounts of computing time and there is a strong incentive to find a more efficient algorithm.

Tidswell et al. [5] and Lucas [6] have devised a more efficient formalism based on the kinematic approximation and this approach has been recently applied to the analysis of organic monolayers on silicon [5] and the photodissolution of silver in chalcogenide glasses [6]. Although, more recently Thomas et al. [8–10] have demonstrated the great value and advantages of the kinematic approximation in the analysis of surfactant monolayer absorption. Its inadequacies are also well known. However the method has attractions for the efficient calculation of reflectivity for systems with a large number if layers, in which absorption and interfacial effects are conveniently included.

In this report we evaluate the extent to which this method can be applied to the study of solid films and make recommendations to its use.

The neutron refractive index at the boundary between two media is defined as

where k_1 and k_0 are the neutron wavevectors, $2\pi/\lambda$, inside and outside the medium. n can be written as

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A comparison of the calculation of neutron reflectivity profiles R(Q) by optical matrix methods and a multilayer method, based on the kinematic approximation, has been made. The latter method provides a fast simple route to calculating R(Q) and may be used with confidence for systems of total thickness $\leq$ 200 Å for a scattering length density difference $\Delta\rho$ of the order of 1.0×10^{-5} Å^{-2} However, it is also clear that refraction at each interface must be included. For a large thicker layers the approximation is poor and the optical matrix method must be used for an accurate evaluation of such reflectivity data over a wide Q range .

1. Introduction

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In this report we evaluate the extent to which this method can be applied to the study of solid films and make recommendations to its use.

The neutron refractive index at the boundary between two media is defined as

$$n = k_1/k_0 \quad (1)$$

where k_1 and k_0 are the neutron wavevectors, $2\pi/\lambda$, inside and outside the medium. n can be written as

$$n = 1 - A\lambda^2 + iC\lambda \quad (2)$$

Where $A = Nb/2\pi$ and $C = N\sigma_{abs}/4\pi$, with N the atomic number density, b the bound coherent scattering length and σ_{abs} the absorption cross-section and λ the incident wavelength. In general the effects of absorption are negligible, and its influence on the reflectivity profiles is described in detail elsewhere [11].

2. The Optical Matrix Method

The optical multilayer matrix methods have been used extensively to evaluate neutron reflection data, and a particularly convenient method is that of Abeles [4]. A characteristic matrix per layer is defined in terms of Fresnel coefficients and phase factors ; derived in optical terms from the relationship between electric vectors in successive layers. the characteristic matrix per layer is then

$$C_M = \begin{bmatrix} \exp(i\beta_{m-1}) & r_m exp(i\beta_{m-1}) \\ r_m exp(-i\beta_{m-1}) & exp(-i\beta_{m-1}) \end{bmatrix} \quad (3)$$

where $\beta m = (2\pi/\lambda)n_m d_m \sin\theta_m$ (optical path length in the film), Fresnel coefficient $r_m = (p_{m-1} - p_m) / (p_{m-1} + p_m)$, and $p_m = n_m \sin\theta_m$. For N layers the elements of the resulting matrix $\mathbf{M_N} = [\mathbf{M_1}][\mathbf{M_2}]...[\mathbf{M_{M+1}}]$ gives the reflectivity

$$R = M_{21}M_{21}^* / M_{11}M_{11}^* \quad (4)$$

It is now possible, following Cowley and Ryan [7] to introduce a roughness or diffuse interface at each boundary of the form originally developed by Nevot and Crocé [12] such that.

$$r_m = \left(\frac{p_{m-1} - p_m}{p_{m-1} + p_m} \right) exp[-0.5(Q_m Q_{m-1} < \sigma >^2)] \quad (5)$$

where $Q_m = 2k\sin\theta_m$ and $<\sigma>$ is the root mean square roughness

3. Kinematic Approximation

Following Als-Nielsen [13] the reflectivity from a real surface can be written in the kinematic approximation in terms of the scattering length density profile normal to the interface

$$R(Q) = R_f(Q)|\phi(Q)|^2 \tag{6}$$

$$\phi(Q) = \int 1/p_\infty (dp/dz) e^{iQz} dz \tag{7}$$

and Rf(Q) is the Fresnel reflectivity for a perfectly sharp interface.

This approximation has been generalised by Tidswell et al. [5] to produce an expression for the reflectivity of N layers with different densities and interfacial widths,

$$R(Q) = R_f(Q)\left|\sum_{i=0}^{N}[(p_i - p_{i+1})/p_o]exp[-iQD_i]exp[-Q^2\sigma^{2i+1}/2]\right|^2 \tag{8}$$

where $\rho_j = N_j b_j$, and $j=0$ represents the substrate (so ρ_o is the density of the substrate), Di is the distance from the semi-infinite substrate ($D_o=0$) to the interface between the ith and $(i+1)$st layers. The thickness of the ith layer is L_i then $D_i = \Sigma L_i$.

The effects of refraction at each interface can also be readily accommodated by substituting for the QD_i term in equation 8 $QD_i = \Sigma Q_j L_j$ where $Q_j = (Q^2 - Q^2_{jc})^{1/2}$ and Q_{jc} is the critical wave vector for the jth layer.

Furthermore, the effect of absorption at each interface can be incorporated by multiplying the terms in sum of equation 8 by the attenuation factor

$$exp[(-\Sigma(N_i\sigma^{abs} L_i/sin\theta)) \tag{9}$$

with $N_i\sigma_1^{abs}$ the atomic number density in layer i multiplied by the neutron absorption cross-section for i.

4. Discussion

Extensive calculations have been made in order to provide a detailed comparison between the two methods of calculation.

Figure 1 illustrates the use of the kinematic approximation to reproduce the calculation of Pershan et al (see figure 4c in reference 5).

In figure 2 it is shown that, providing the region of total reflection is avoided, the kinematic approximation agrees well with the optical matrix method for the reflectivity from the interface between two bulk media at higher values of Q.

In figure 3 the validity of the kinematic approximation is explored for different film thicknesses and for a constant contrast between the film and substrate. For thicknesses < 100Å the agreement with the optical matrix calculation is good over a wide Q range. For a thickness of 100Å there is already some discrepancy at lower values of Q (see figure 3c). For thicknesses > 100Å the discrepancy becomes progressively larger and extends to higher Q values: such that for films > 500Å the deviations are most marked.

In figure 4 the effect of varying the contrast between the film and substrate for a fixed film thickness is explored. For a thickness as great as 500Å there can still be reasonable agreement between the kinematic approximation and the optical matrix calculation provided the contrast between the film and substrate is small (ie the film is a small perbutation).

In figure 5 the effect of not including refraction between the layers is investigated. It is clearly important in the kinematic approximation to include the effects of refraction (see figures 5 b,c for the case of multilayer structures). Furthermore it is important to note in this case that the kinematic approximation agrees well with the exact calculation in the region of the first order Bragg peak for these multiple bilayers, and only deviates at lower Q in the region of total reflection.

In the kinematic approximation the effects of adsorption can be included approximately in the form of a attenuation factor (see equation 9), whereas in the exact optical matric calculation it is included as the imaginary component of the refracture index. As described in detail elsewhere (11) the interference pattern is changed as the adsorption cross-section increases and the effect is most prominent in the region of total reflection. Hence it is difficult to make a meaningful comparison in this region of Q using the kinematic approximation.

5 . Summary

We have made a comparison of reflectivity profiles calculated by the full optical matrix and kinematic approximation methods. The kinematic approximation provides a rapid and simple way to calculate R(Q) profiles, but can only be used with confidence for systems with relatively small number of thin layers. It is important to include the effects of refraction in the calculation. It is evident that the inclusion of adsorption in an effective attenuation factor is not in general a good approximation. For more complicated systems, it is necessary to use the existing optical matrix method [3]. Within these restrictions the kinematic method will enable R(Q) profiles to be calculated in 1/4 to 1/5 the cpu time of the corresponding optical matrix method.

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Figure Captions :

Legend: Nbs,a - scattering length density of substrate, air ; $\sigma_{l,a}$ surface roughness at film/substrate and film/air interface. d_l layer thickness in Å. No instrumental effects included ie. $\Delta\theta = 0$.

1) Comparison with previous work. $|\phi(Q)|^2$ vs. Q for a system of three discrete layers on a substrate (see ref 5 figure 4c and table 1). Refraction effects included. 0 (substrate) - crystalline Si; 1 - SiO_2, 2 - interface; 3 - alkyl tails; 4 - air. $d_1 = 16.8$Å, $d_2 = 0.7$Å, $d_3 = 23.5$Å. $Nb_1 = 1.895 \times 10^{-5}$ Å^{-2}, $Nb_2 = 2.467 \times 10^{-5}$ Å^{-2}, $Nb_3 = 0.848 \times 10^{-5}$ Å^{-2}. $\sigma_{01} = 1.0$ Å, $\sigma_{12} = 1.0$Å, $\sigma_{23} = 3.2$ Å, $\sigma_{34} = 2.4$ Å.

2) R(Q) profiles for a single sharp interface. $\theta = 1.5°$ $\lambda = 0.5 - 15$Å Nbs = Nbl = 0.21x10-5 Å-2 Nba = 0 ;$\sigma_l = \sigma_s = 0$. kinematic (+), matrix (-).

3) Effect of varying layer thickness on R(Q) at constant contrast. Refraction effects included. $\theta = 1.5°$ $\lambda = 0.5 - 15$Å Nbs = 0.21×10^{-5} Å^{-2} Nbl = 0.50×10^{-5} A^{-2} Nba = 0 ;$\sigma l = \sigma s = 0$. kinematic (+), matrix (-).
   a) dl = 20Å
   b) dl = 40 Å
   c)(i) dl = 100Å and (ii) discrepancies at low Q
   d)(i) dl = 200Å and (ii) low Q
   e) dl = 500Å
   f) dl = 1000Å.

4) Effect of varying contrast at constant layer thickness on R(Q). Refraction effects included. d = 500Å, Nbs = 0.21×10^{-5}Å^{-2} $\sigma = \sigma s = 0$. $\lambda = 0.5 - 15$Å, $\theta = 1.5°$. kinematic (+) matrix (-)
   a) Nbl/Nbs = 0.5
   b) Nbl/Nbs = 0.95
   c) Nbl/Nbs = 1.5
   d) Nbl/Nbs = 2.83 see also figure 3 e

5) Effect of refraction at each interface on R(Q) as calculated by kinematic method. $\theta = 1.5°$ $\lambda = 0.5 - 15$Å Nbl = 0.50×10^{-5} Å^{-2} Nbs = 0.21×10^{-5} Å^{-2} Nba = 0

   Single layer
   a) dl = 500 Å ;$\sigma l = \sigma s = 0$ (+) refraction included (-) no refraction effects. see also figure 3e

   10 x bilayers $d_1 = d_2 = 50$Å $\sigma_1 = \sigma_2 = \sigma a = 5$Å Nba = 0.0
   kinematic refraction included (+) kinematic no refraction effects (Δ) matrix calculation (-)
   b) $Nb_1 = 0.1 \times 10^{-5}$ Å^{-2}, $Nb_2 = 1.0 \times 10^{-5}$ Å^{-2}
   c) $Nb_1 = 0.5 \times 10^{-5}$ Å^{-2}, $Nb_2 = 0.4 \times 10^{-5}$ Å^{-2}

computing time for b) and c) kinematic calculation is a factor of two faster than matrix calculation.

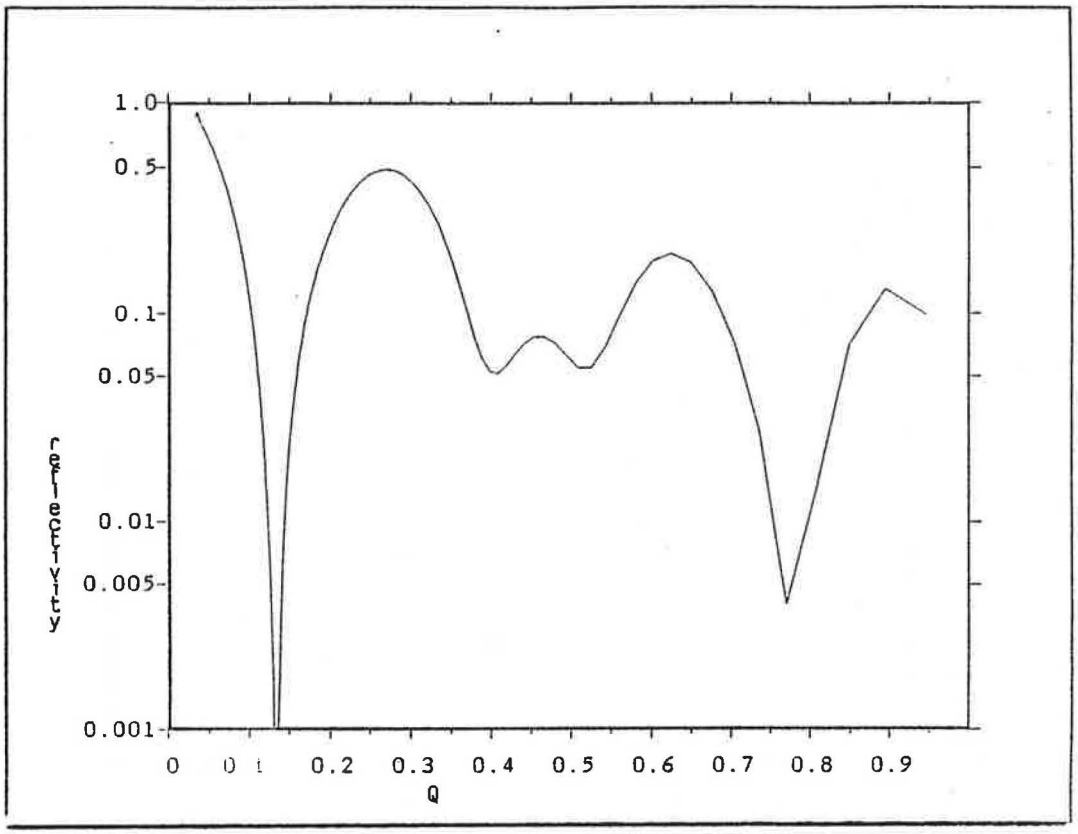


Figure 1

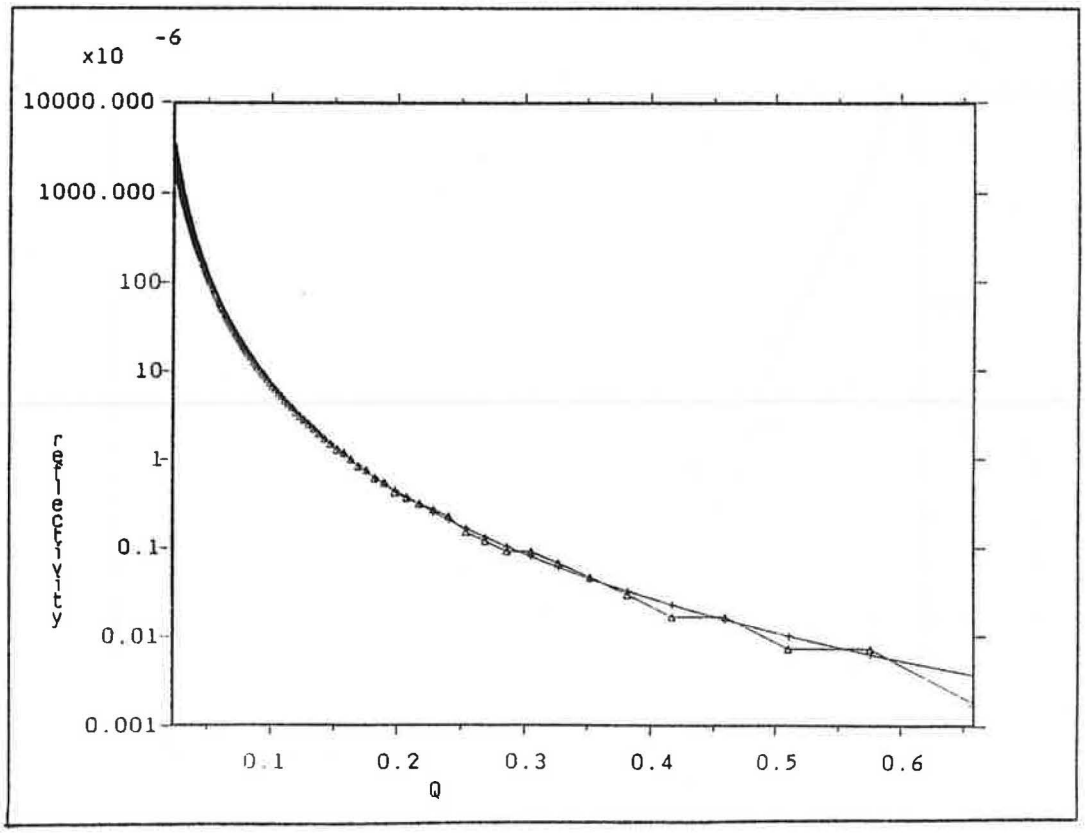
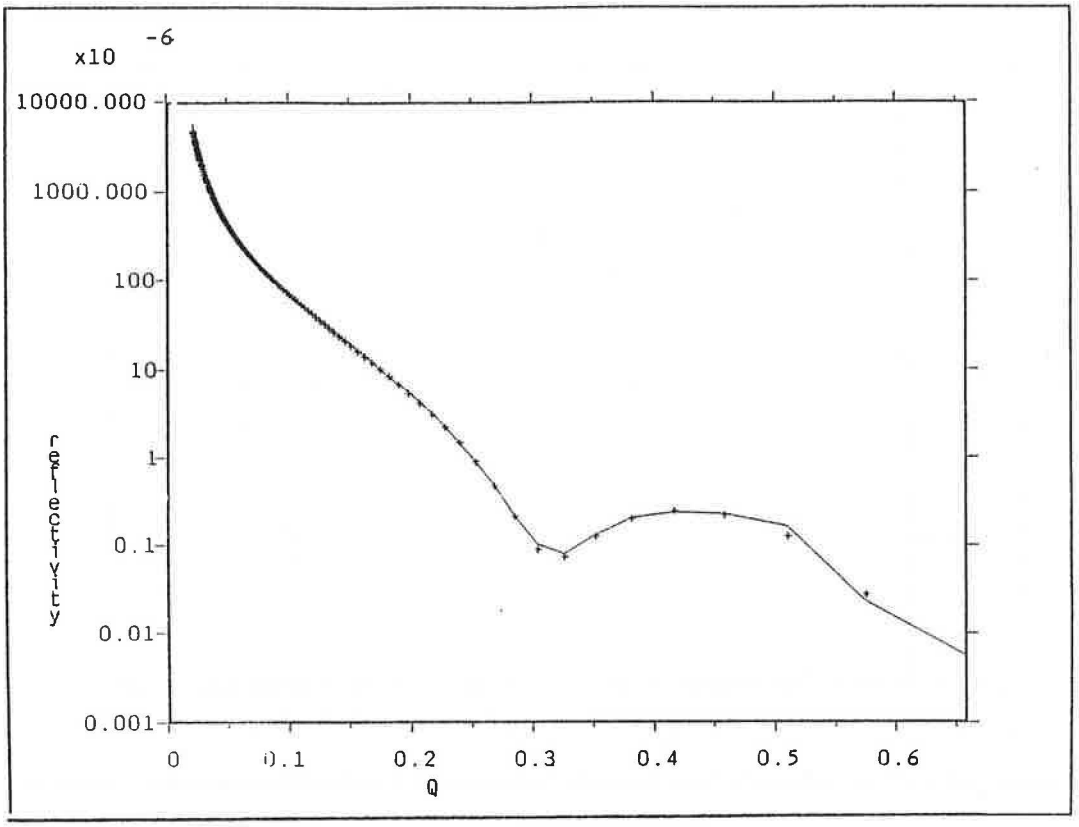


Figure 2



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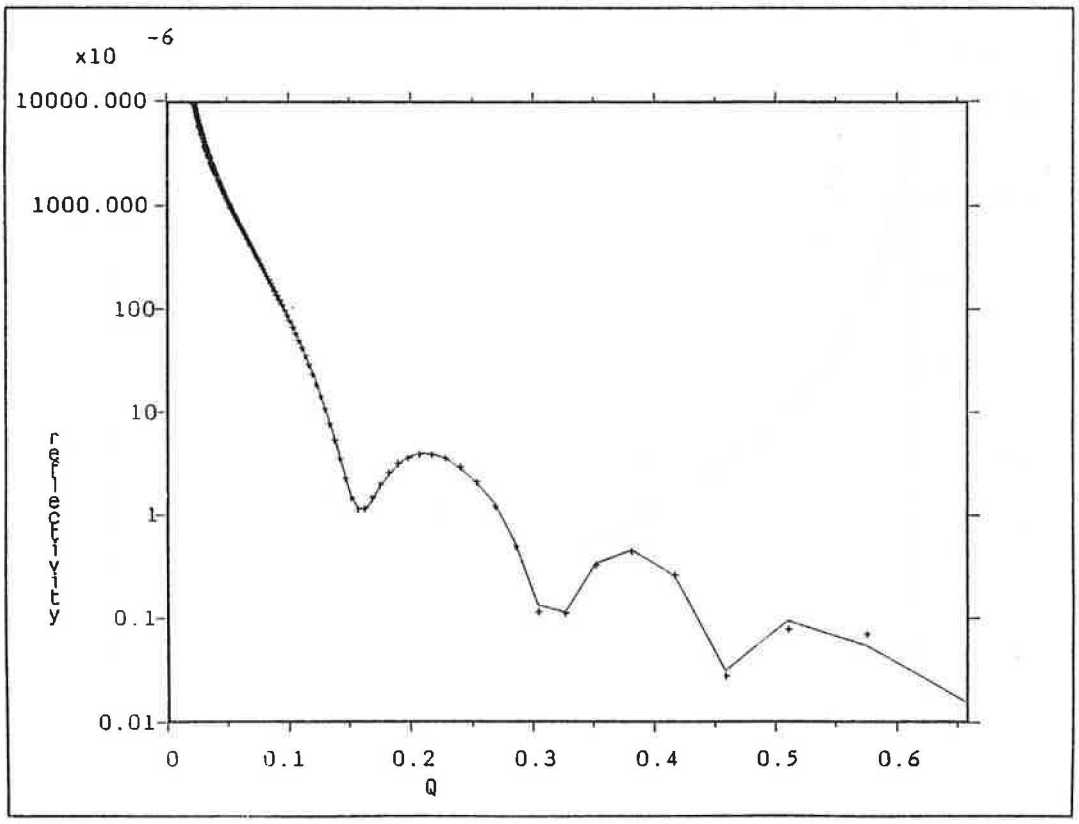

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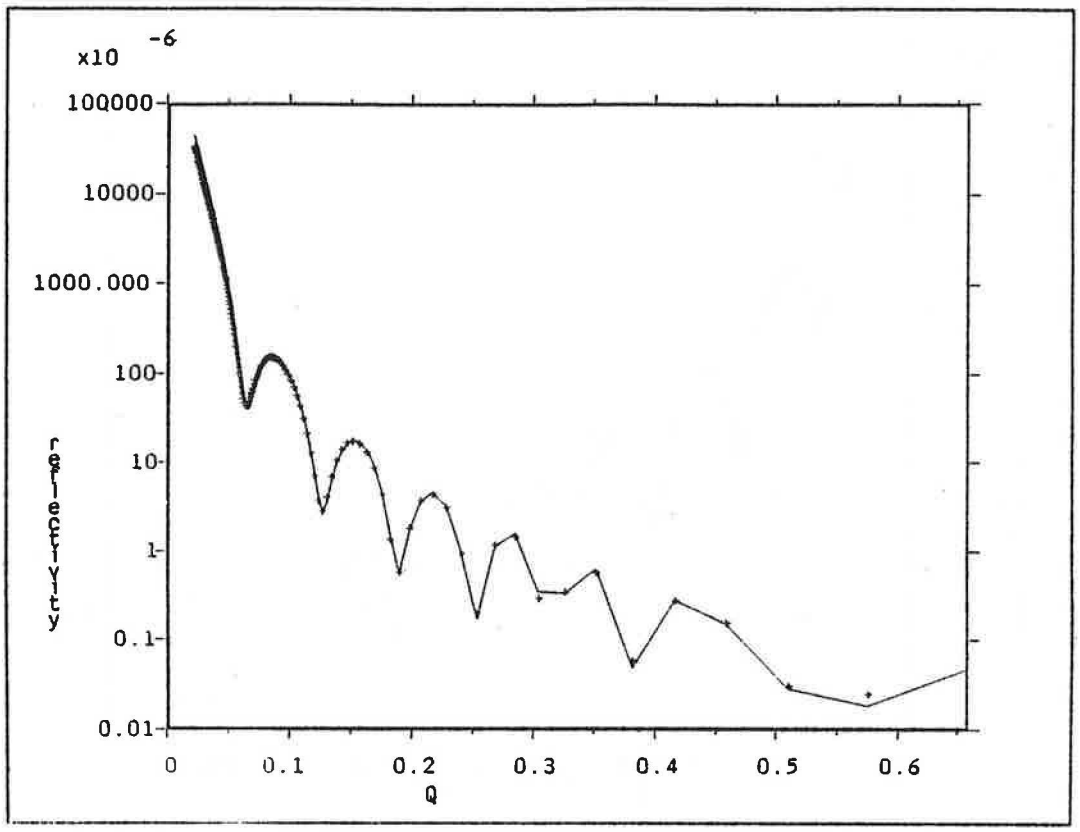
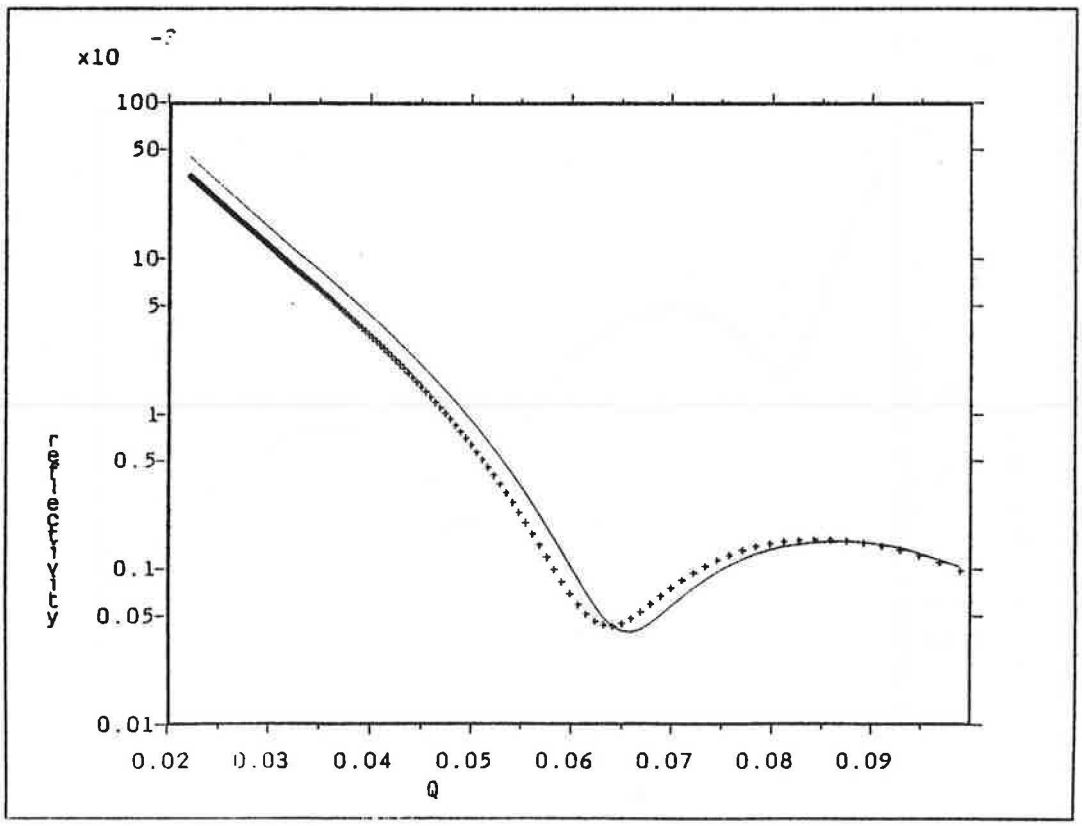


Figure 3(c)(i)



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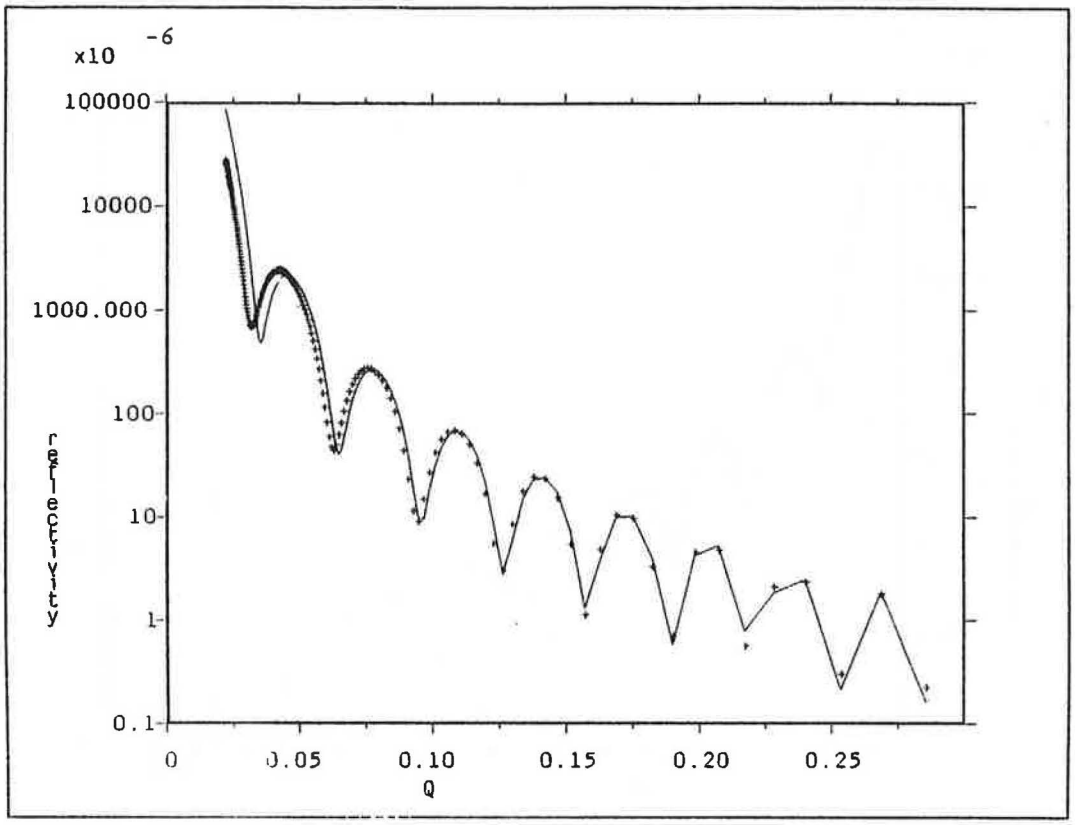

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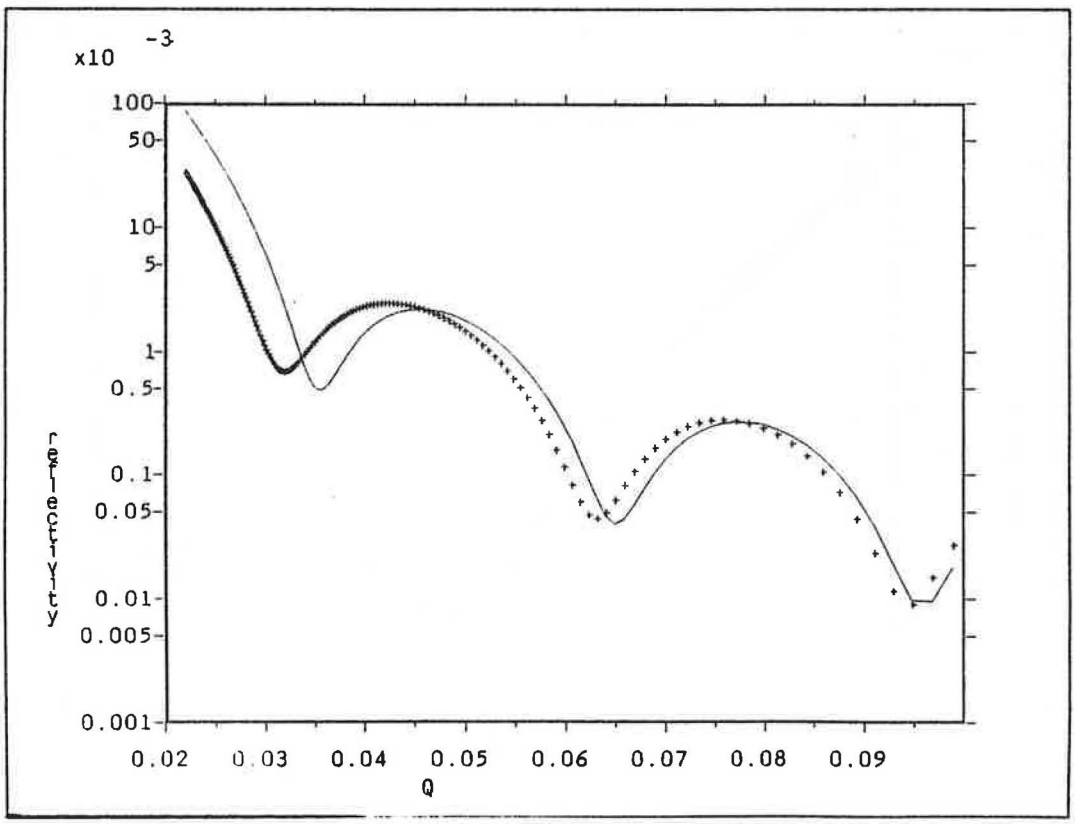

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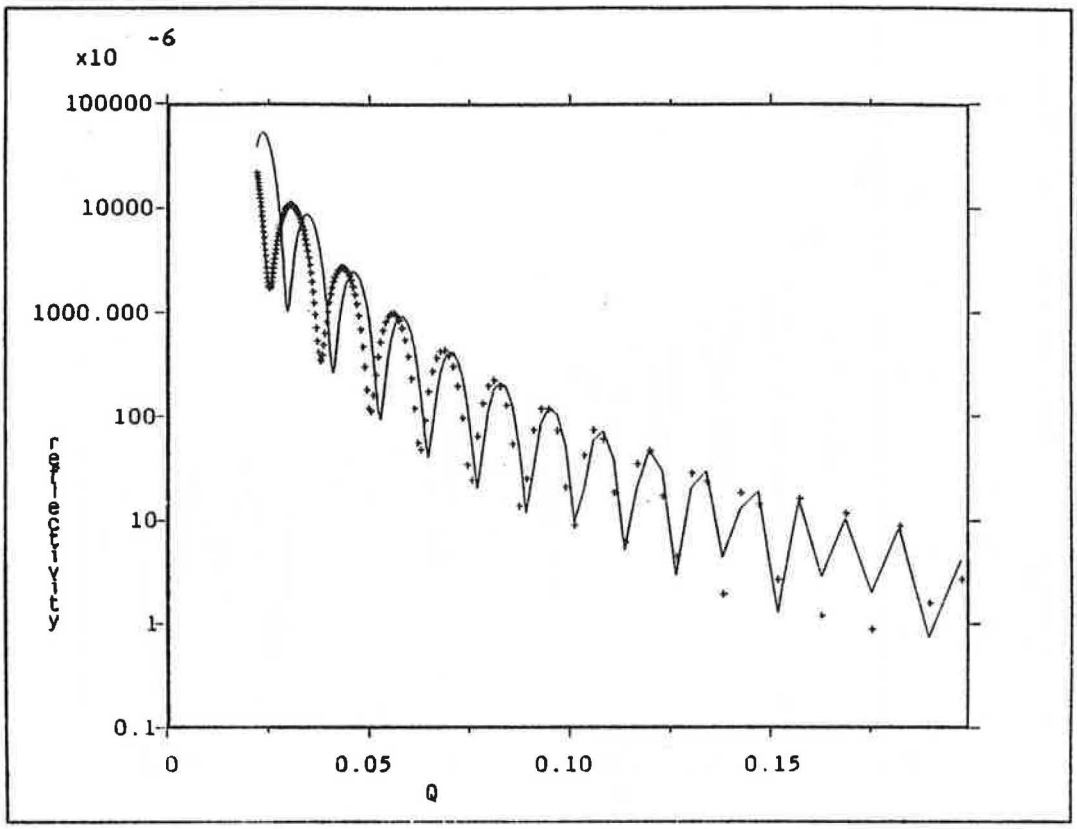

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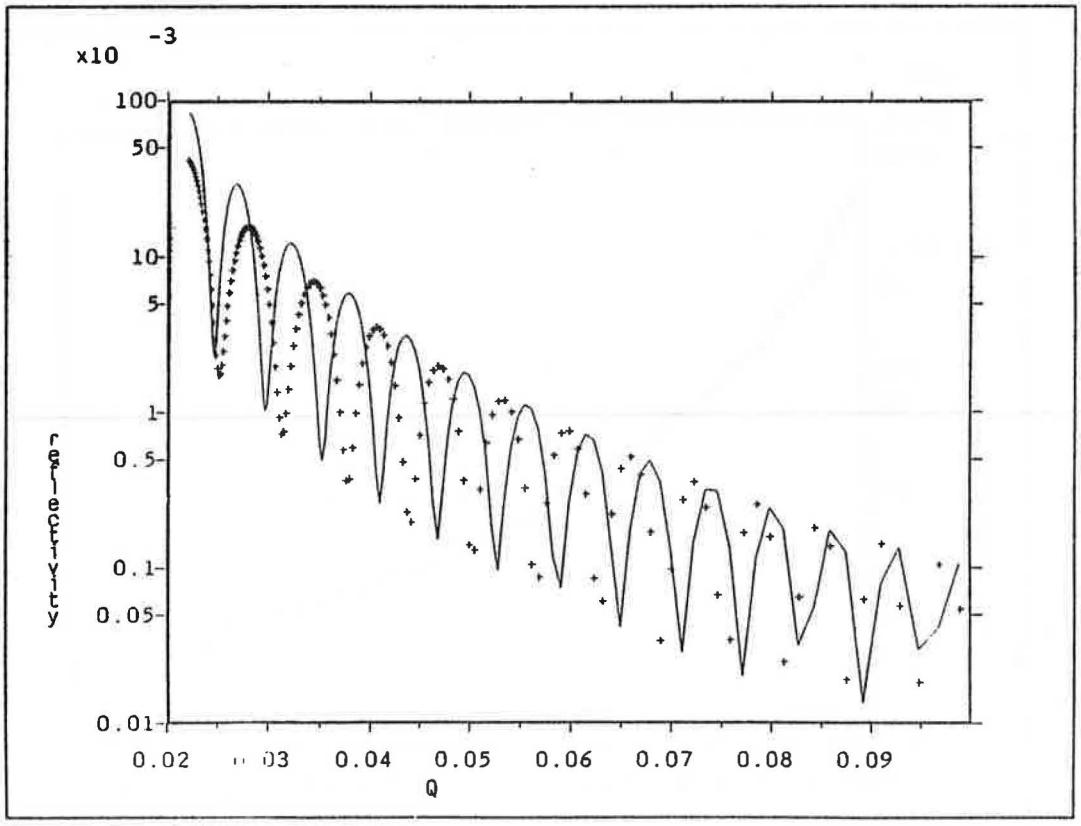

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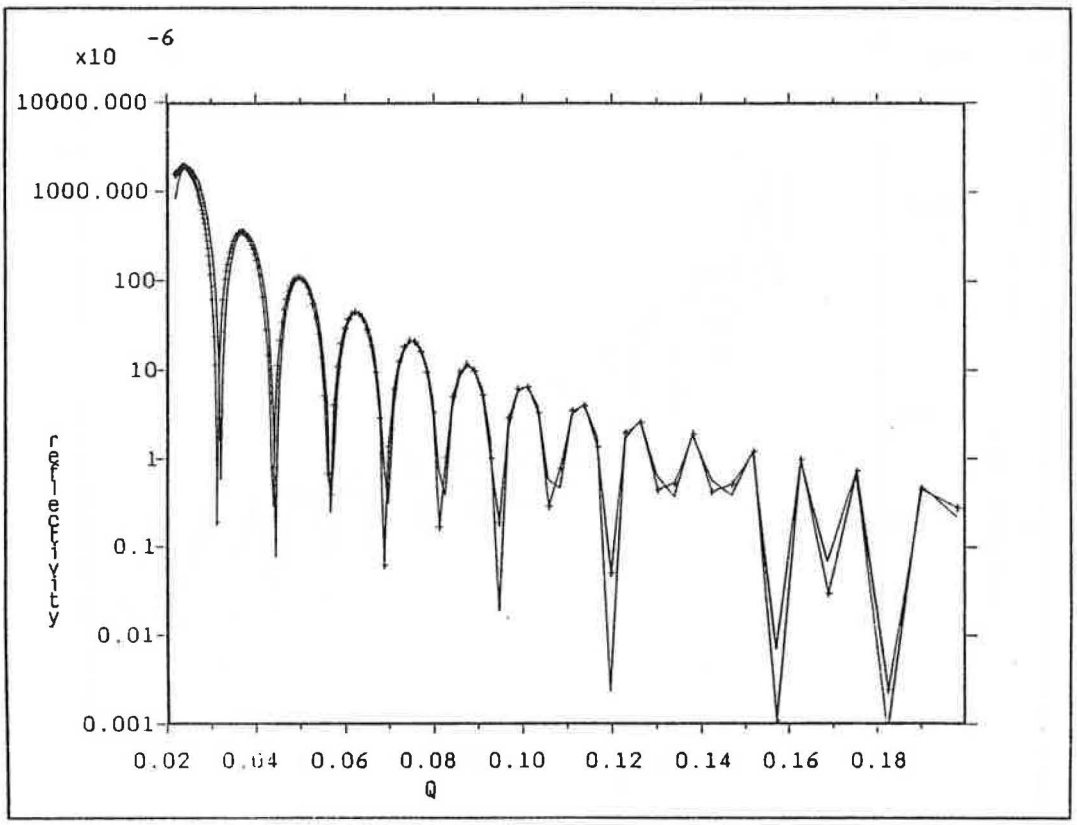

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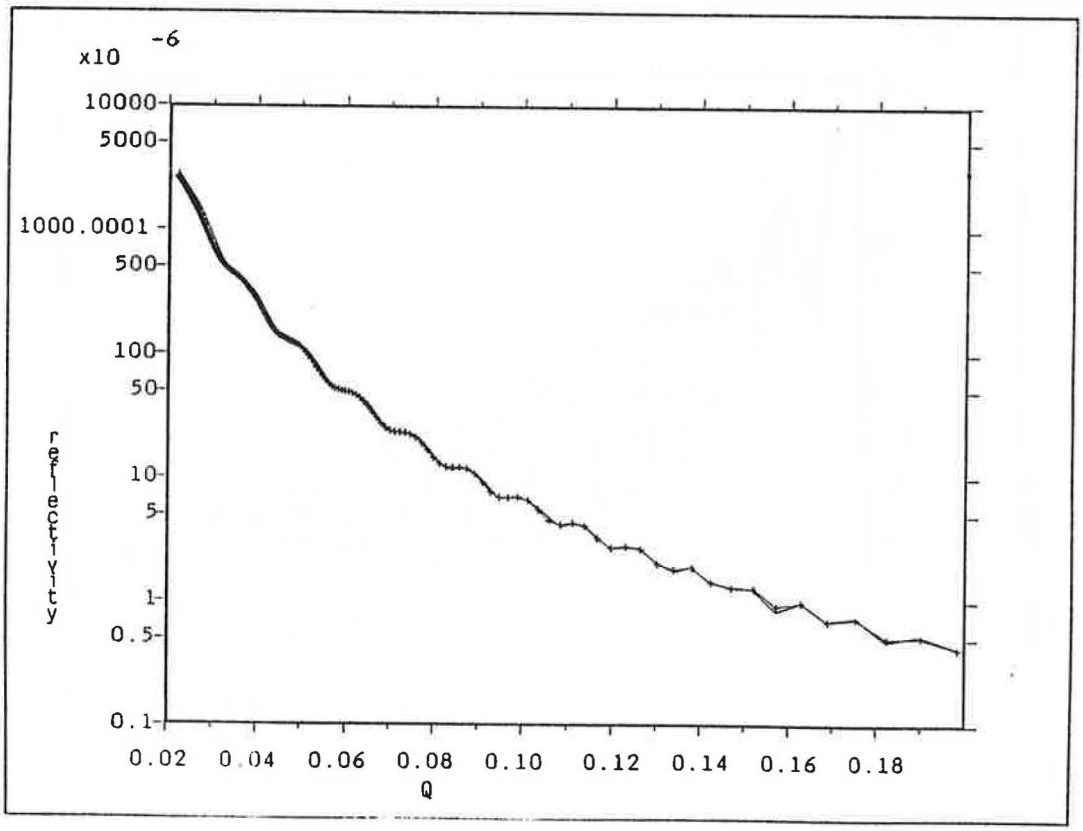

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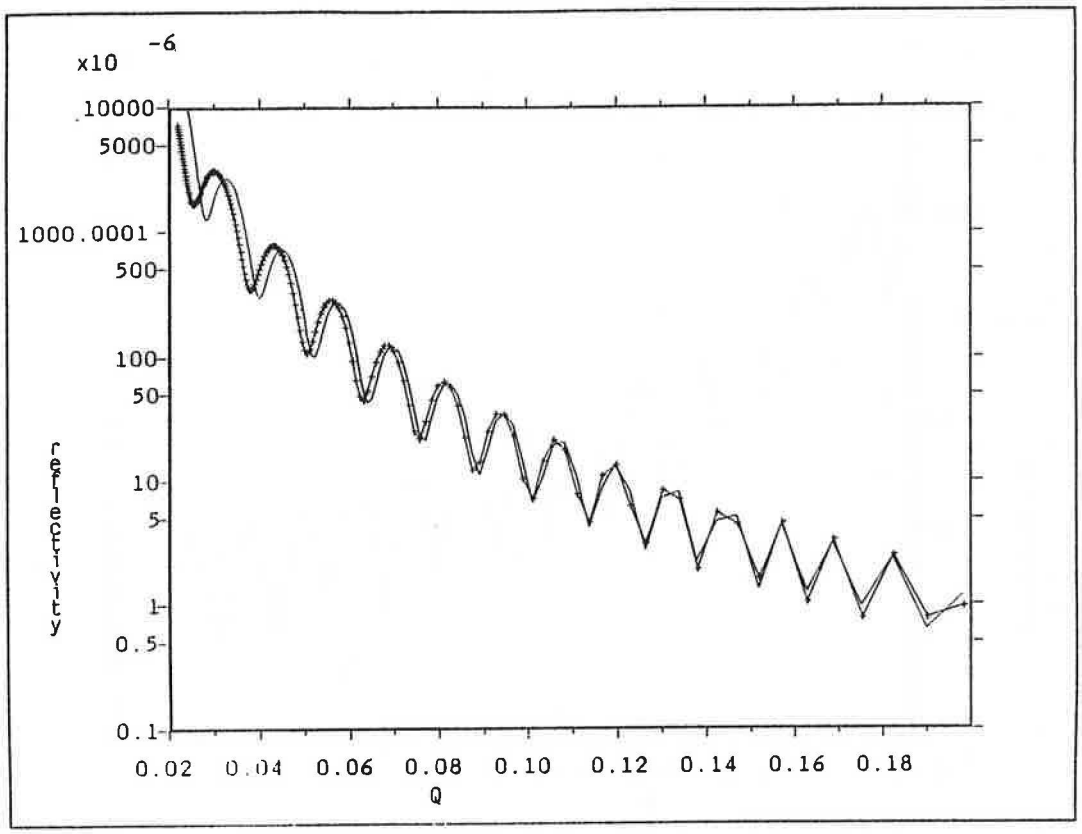

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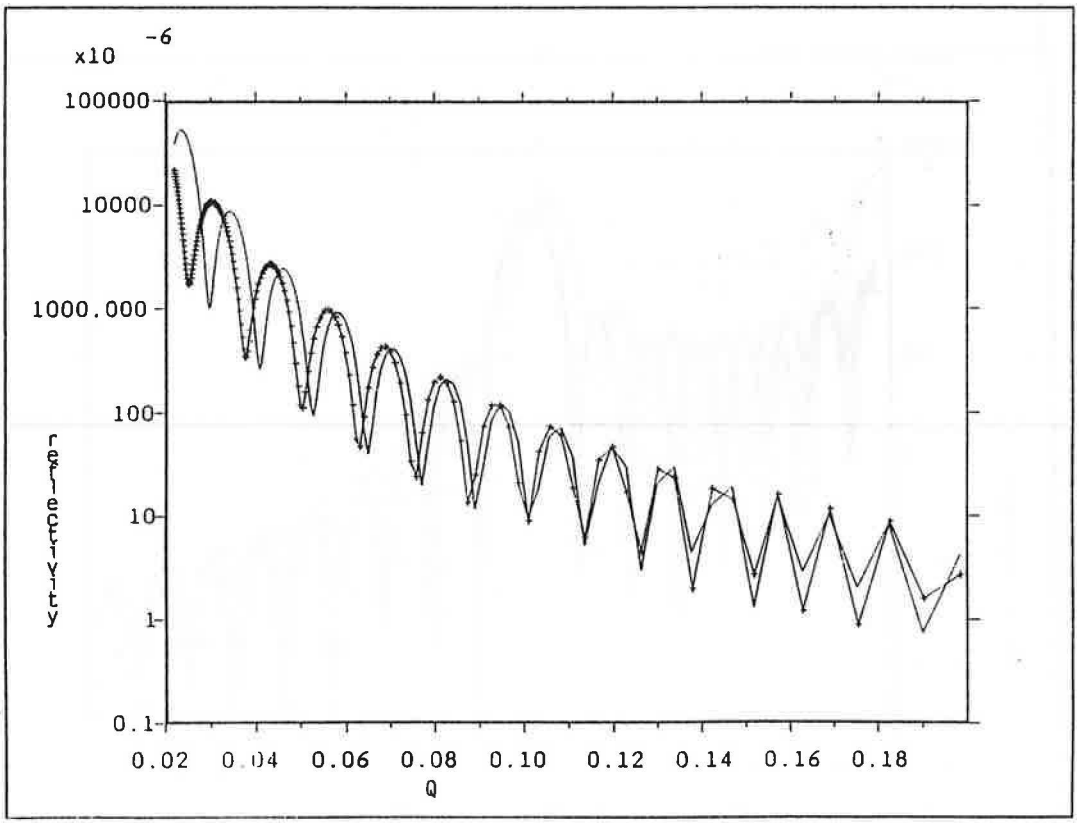

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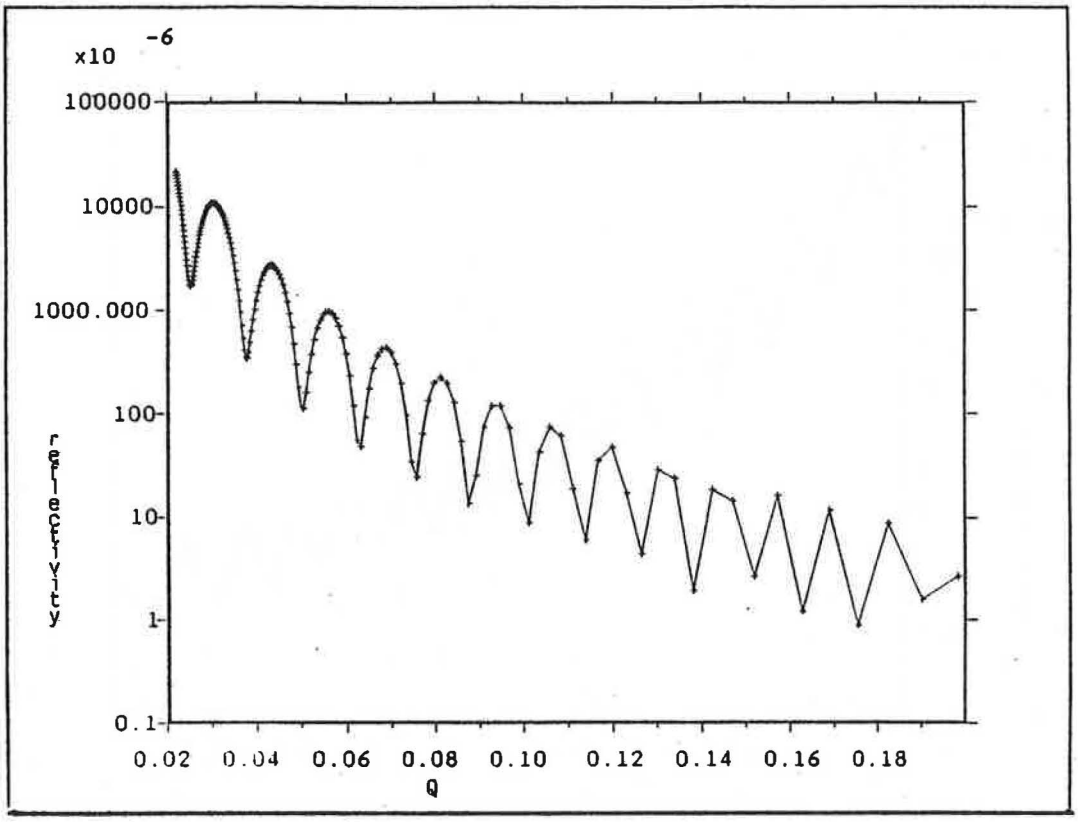

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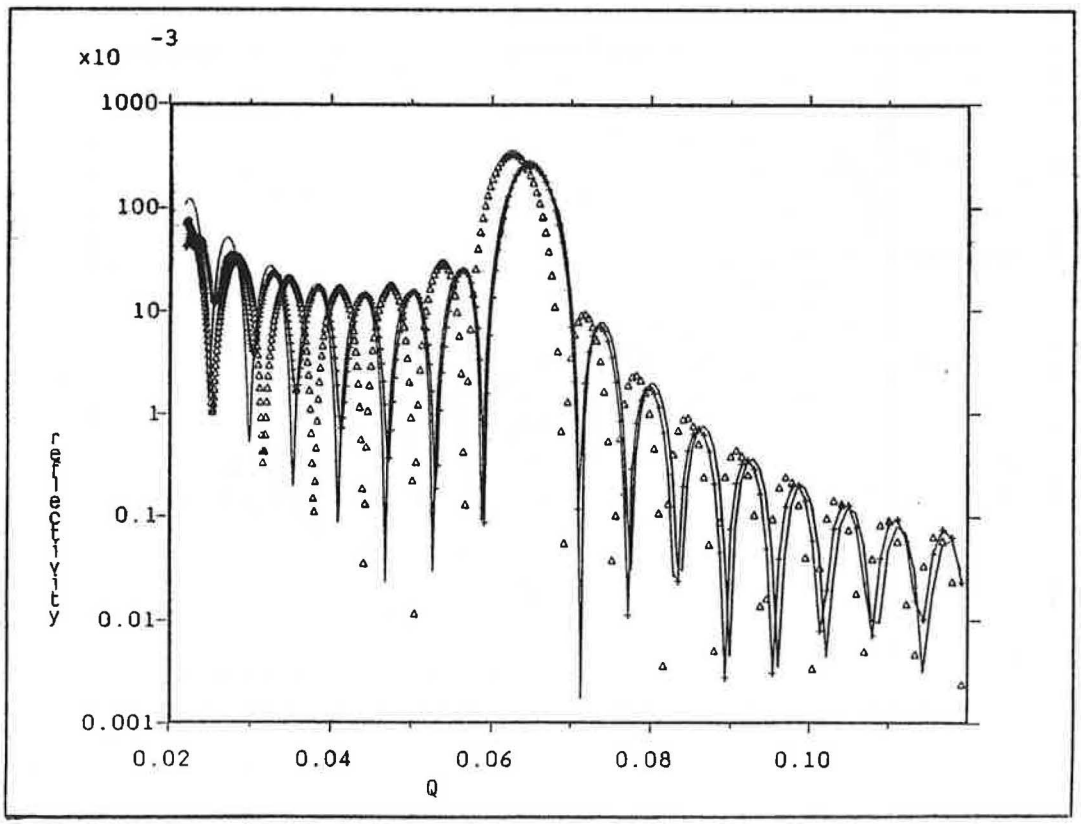

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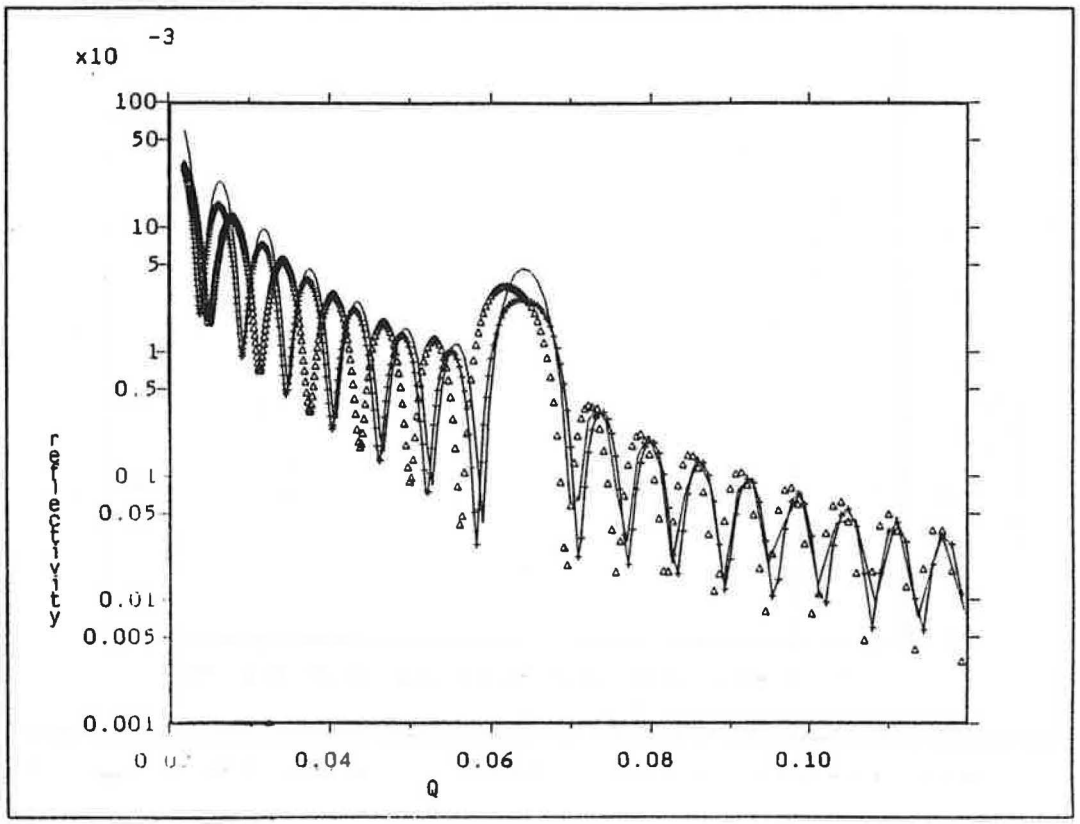
x10 -3
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1
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Q

Figure 5(c)

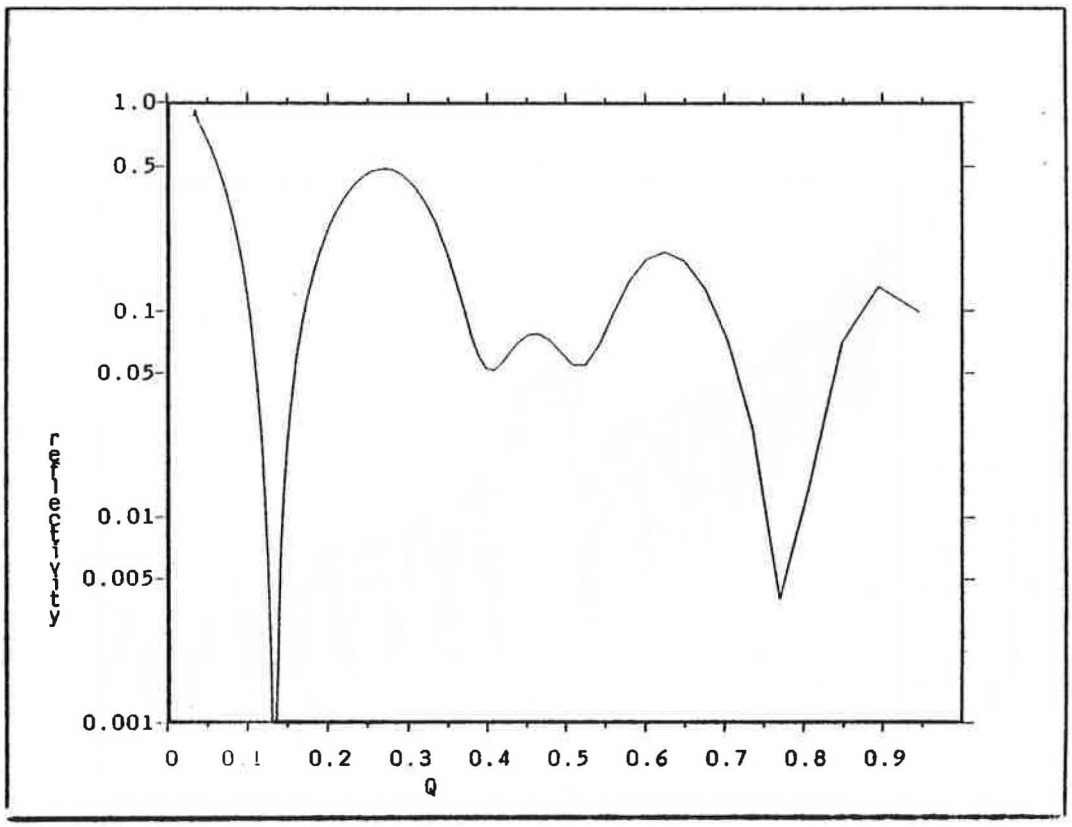
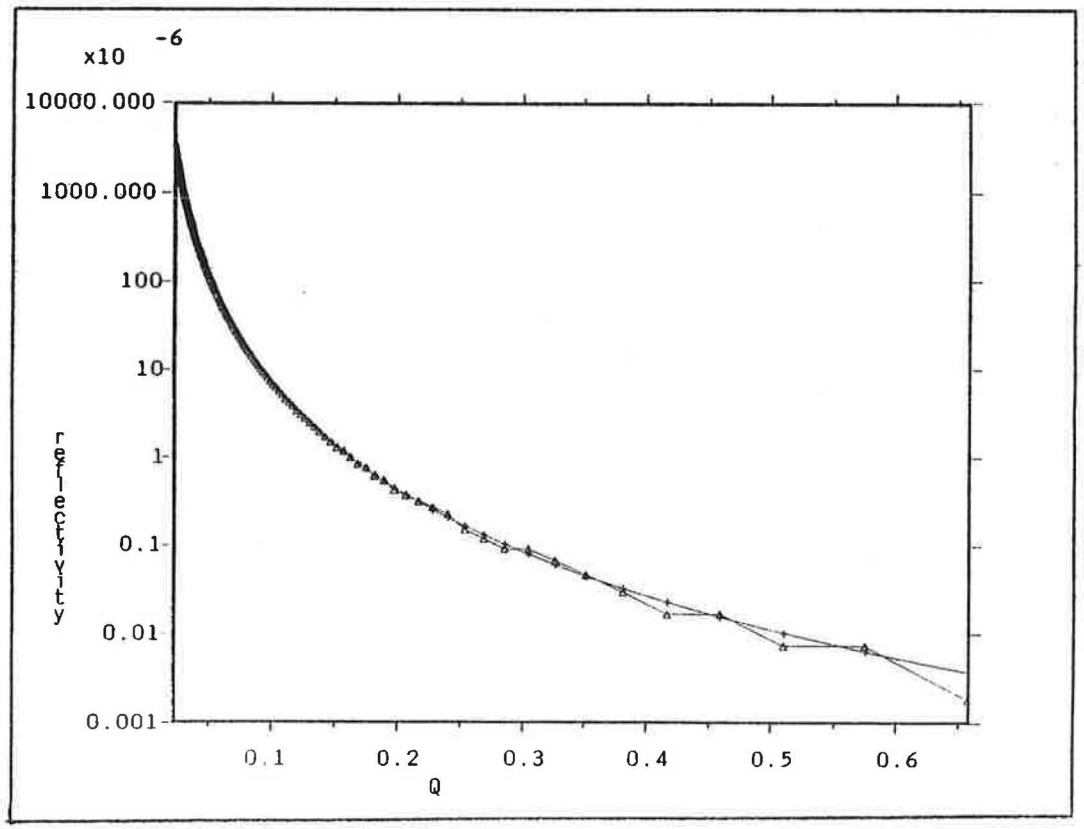


Figure 1



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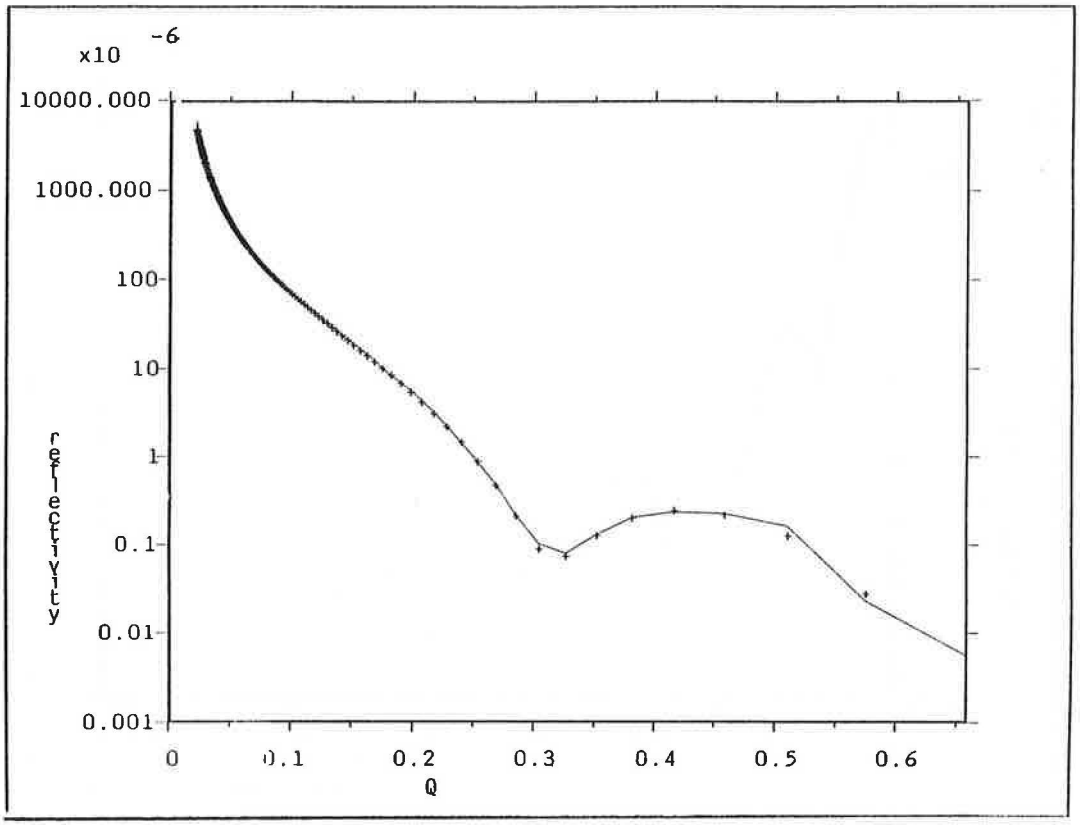
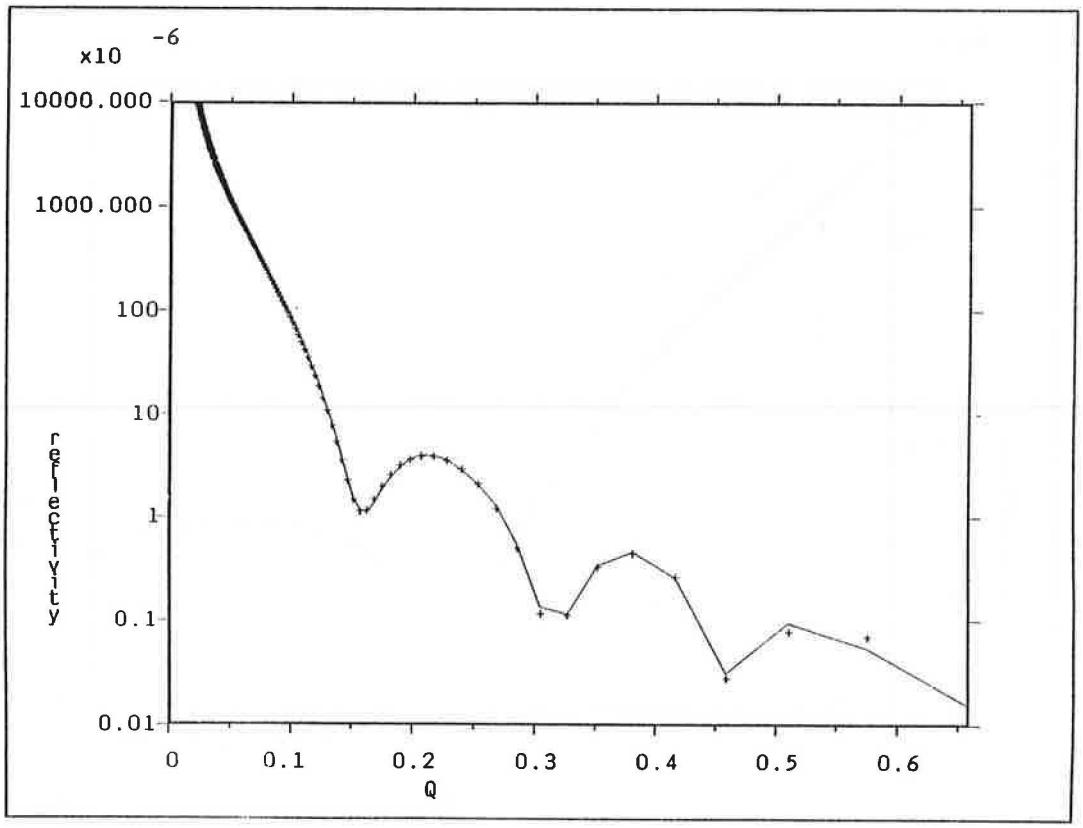


Figure 3(a)



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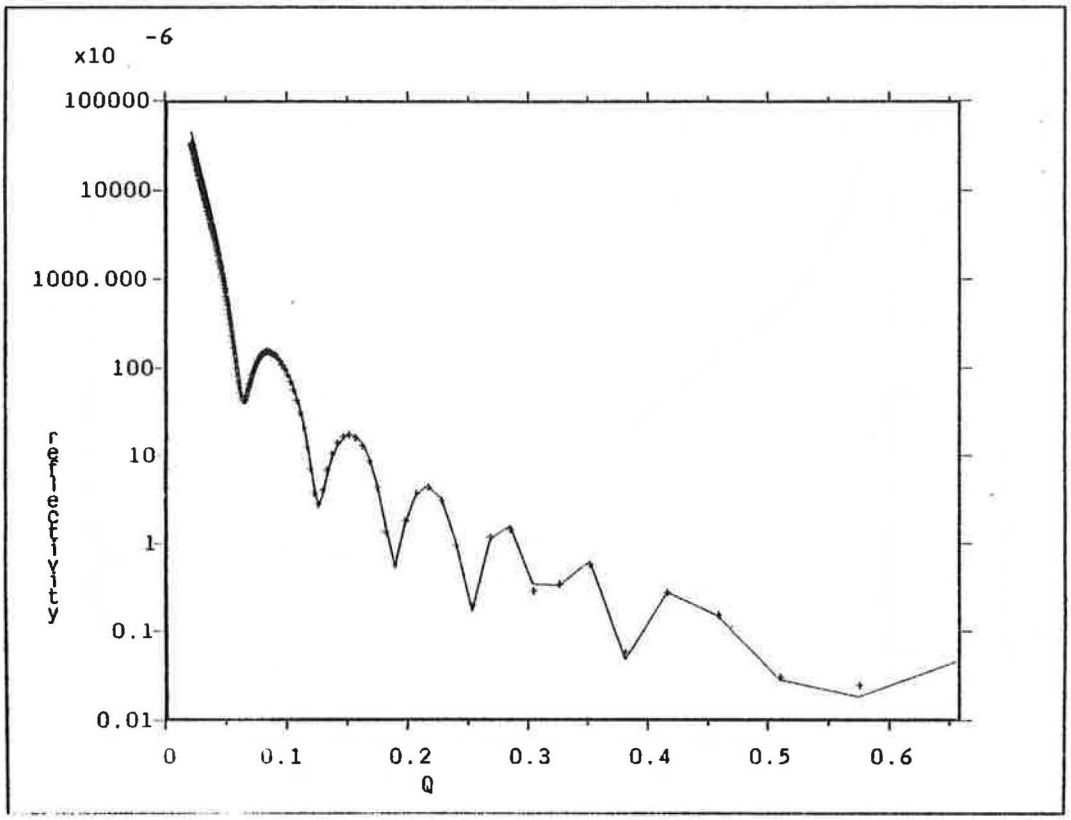
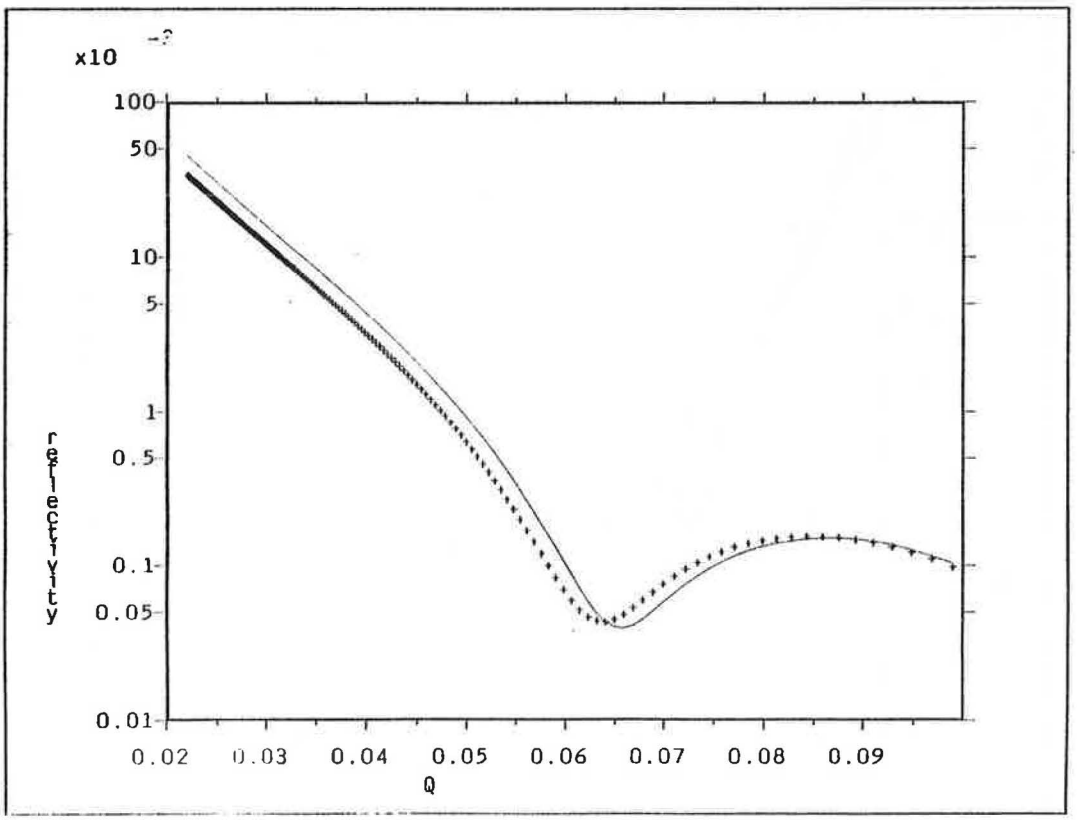


Figure 3(c)(i)



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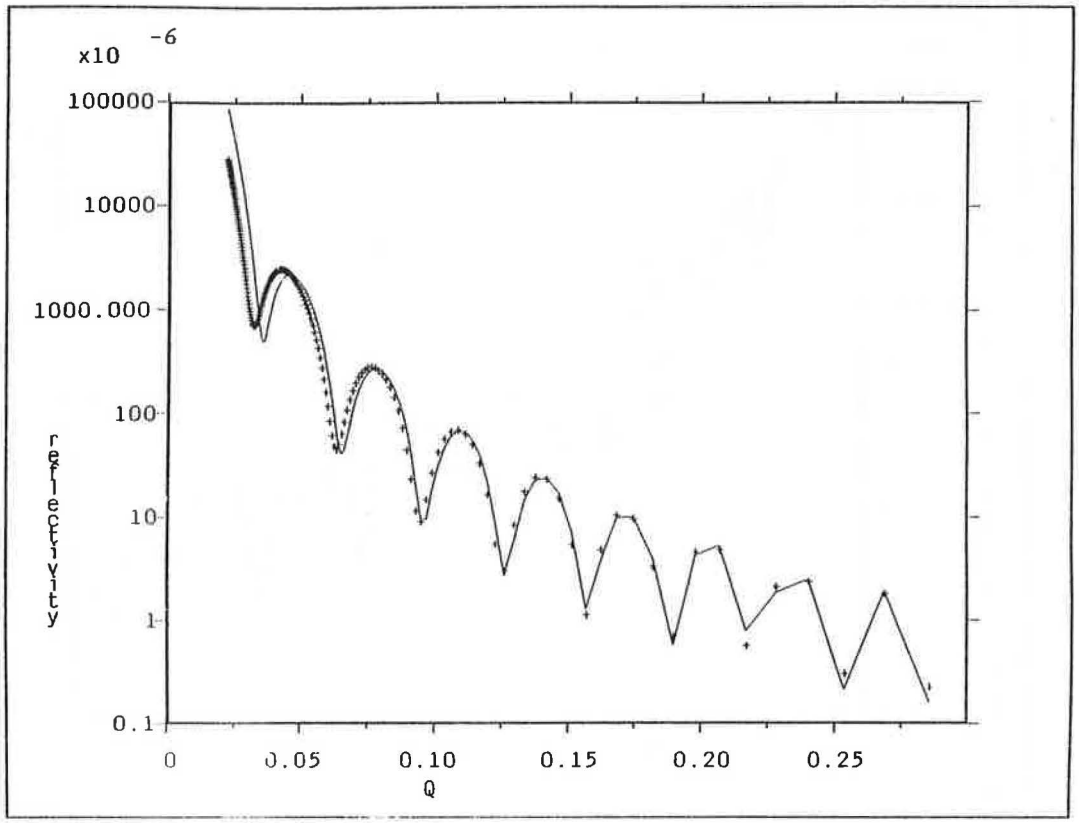

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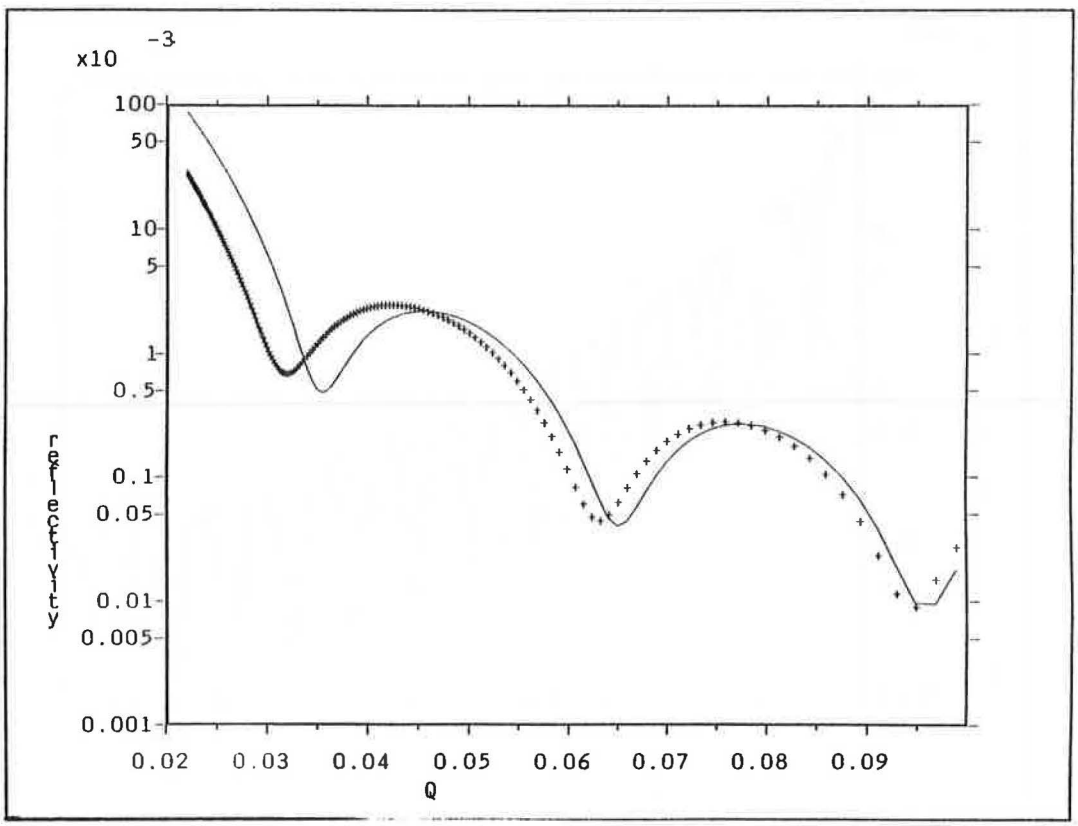

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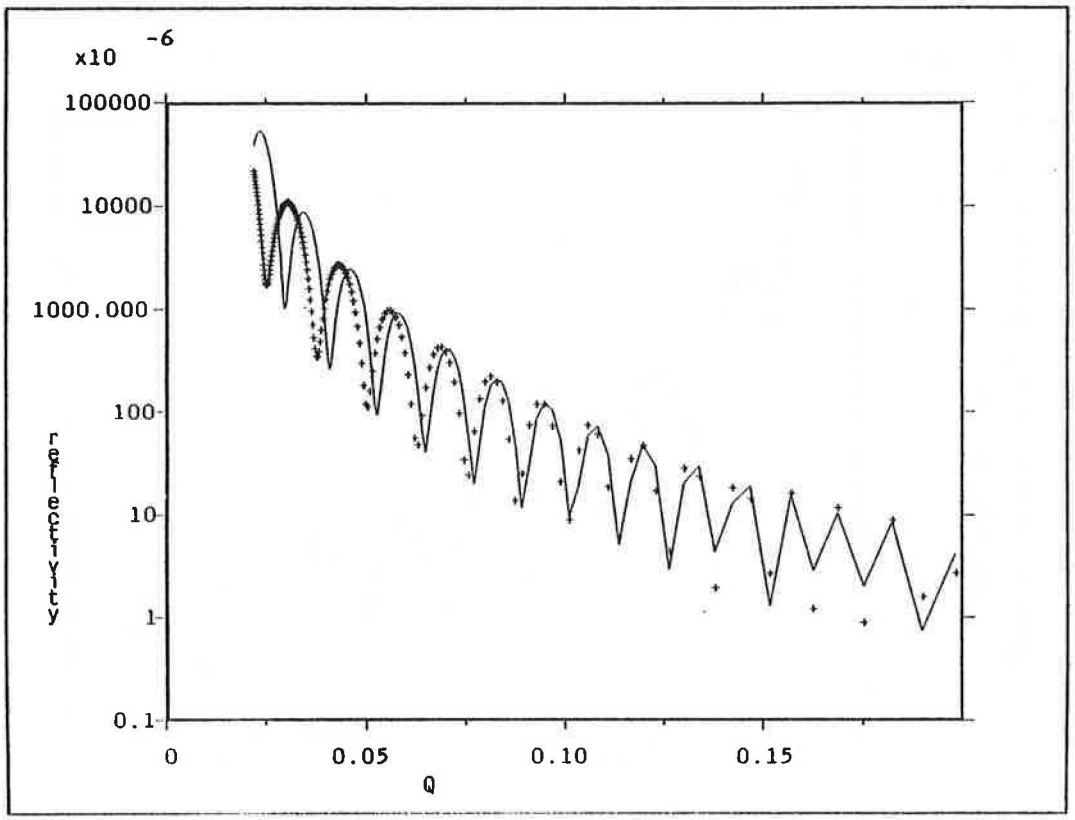


Figure 3(e)

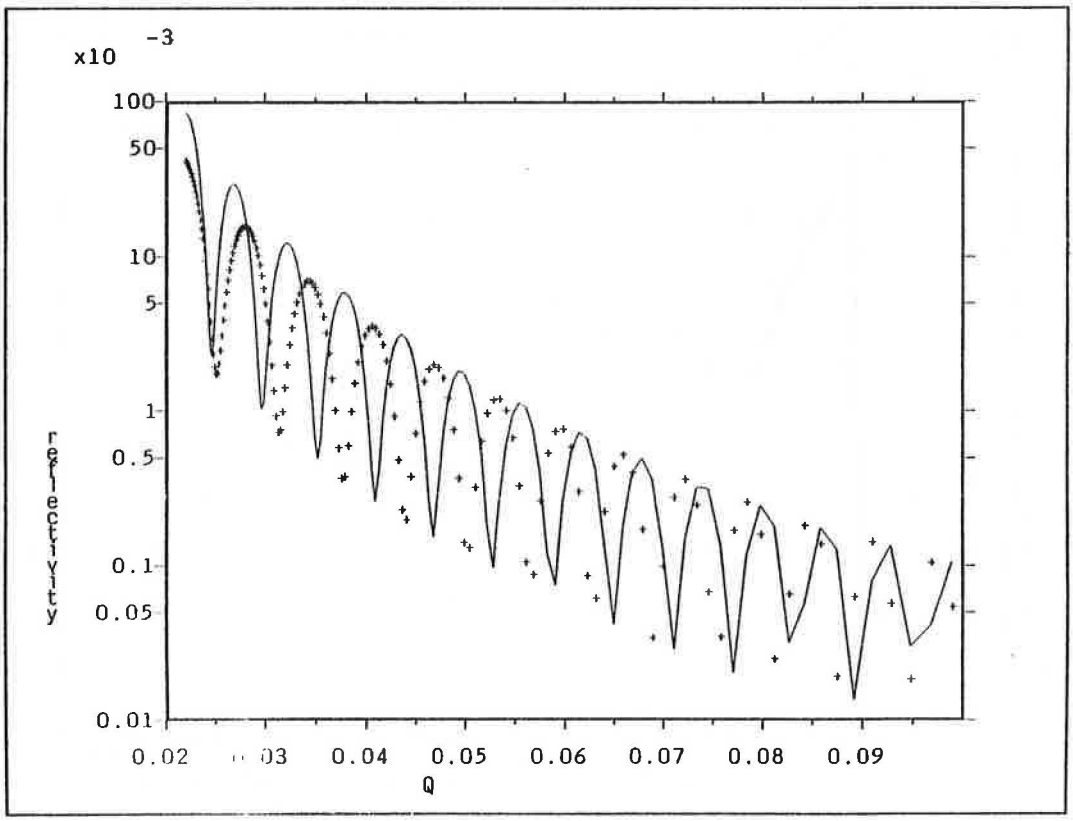


Figure 3(f)

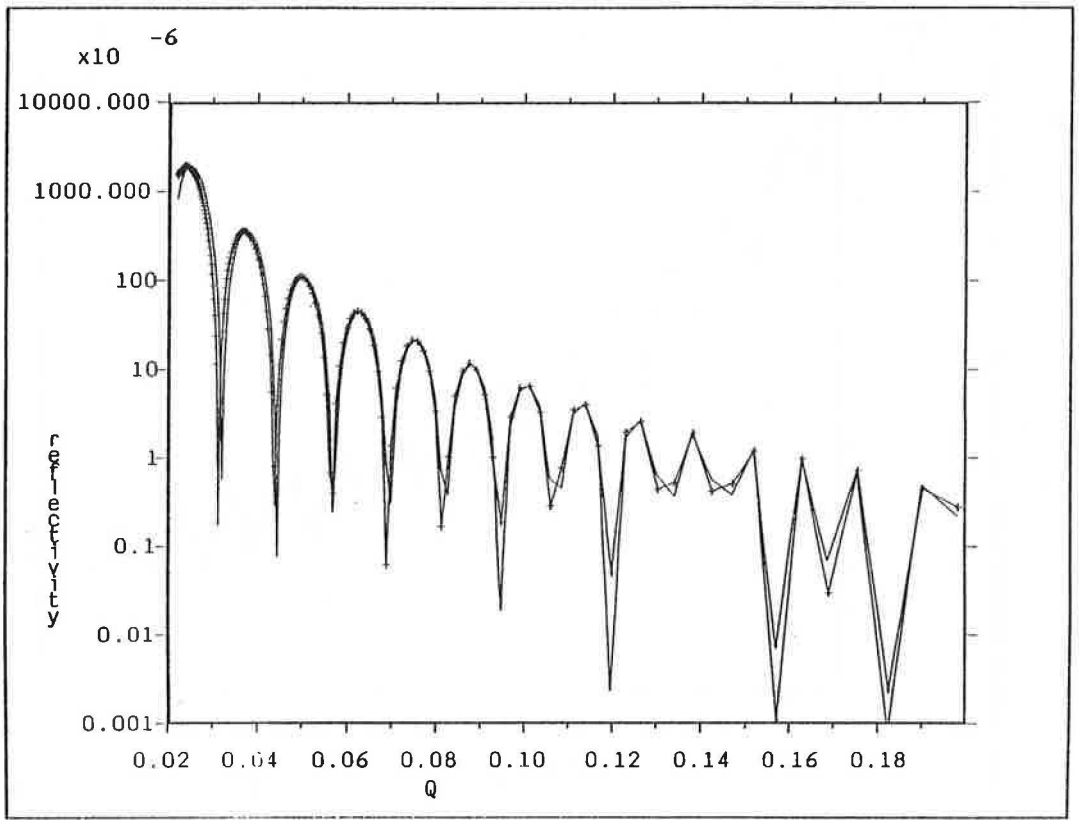


Figure 4(a)

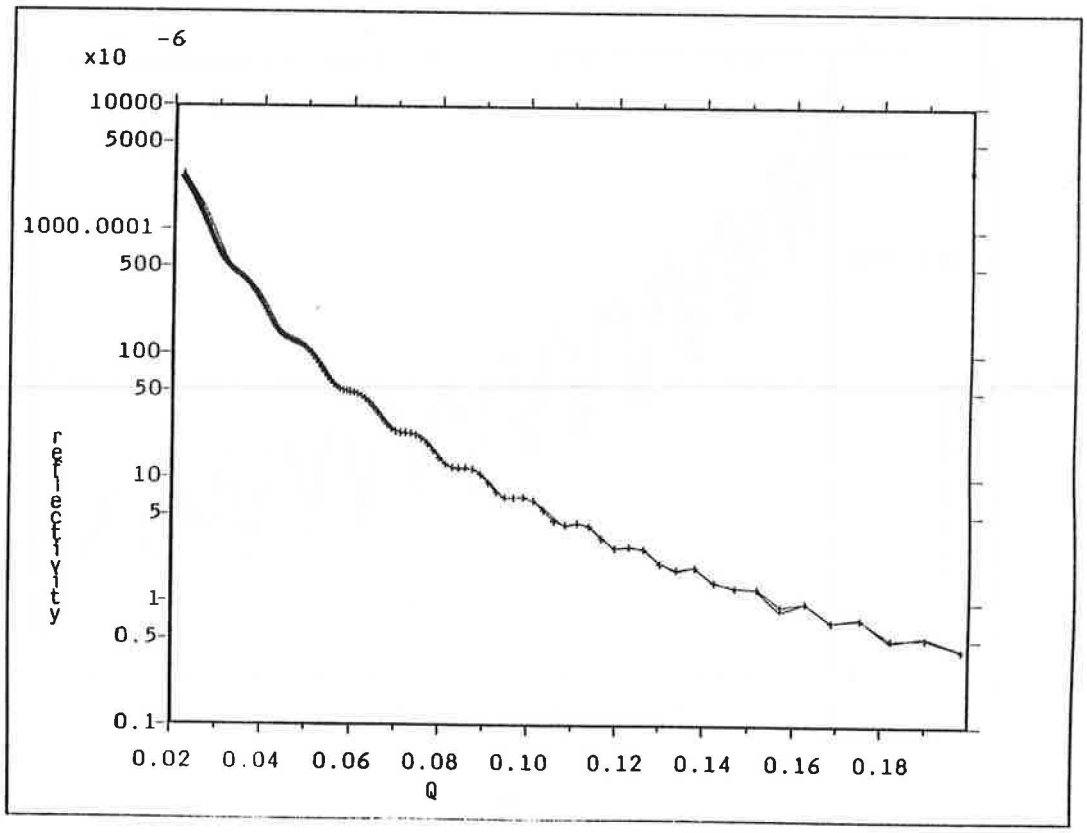


Figure 4(b)

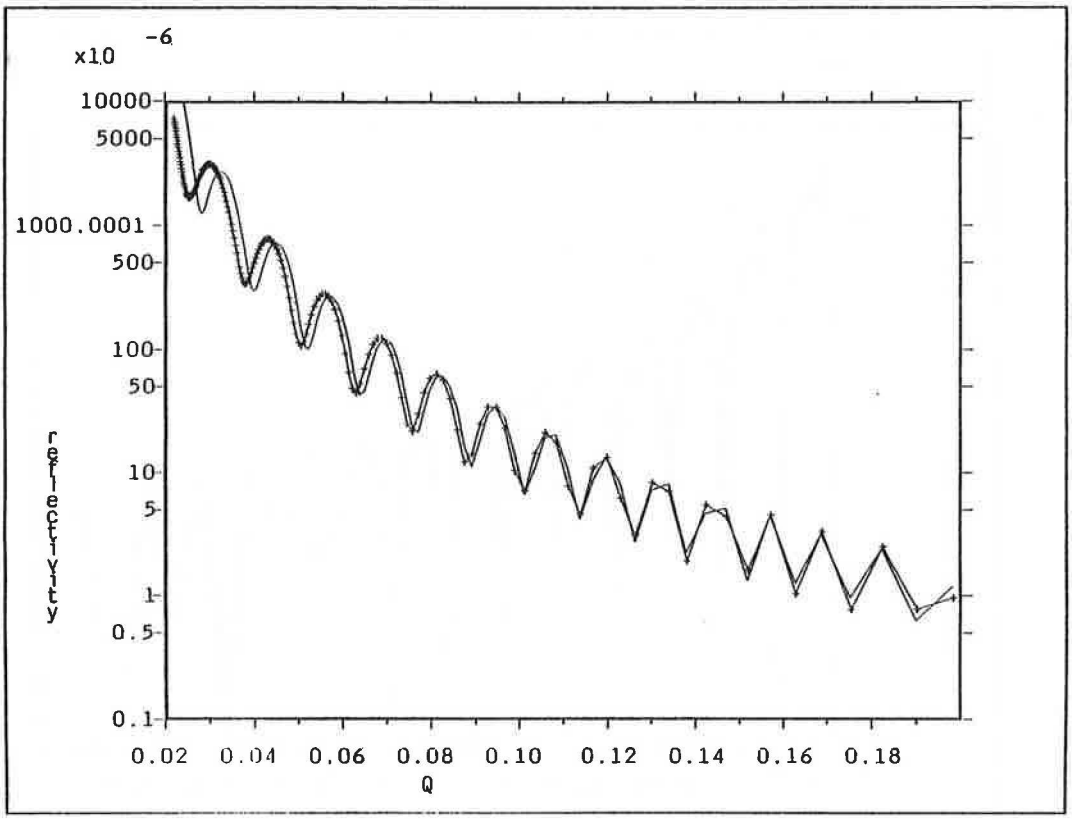


Figure 4(c)

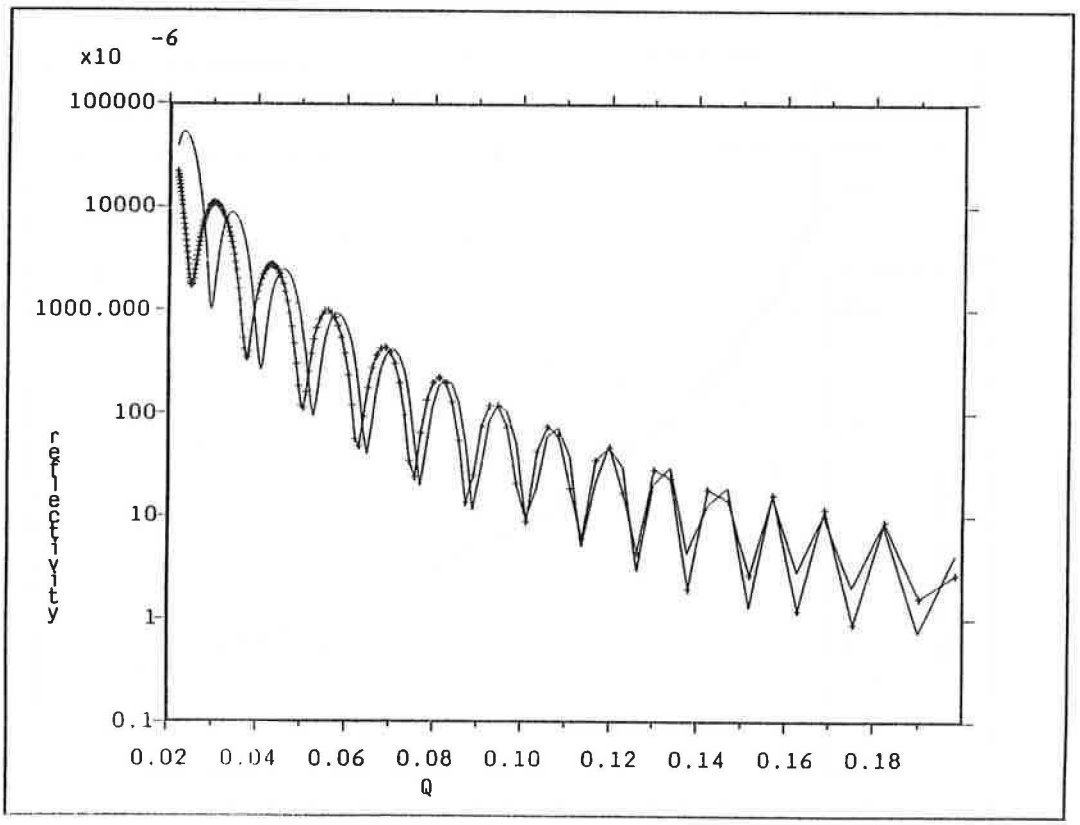


Figure 4(e)

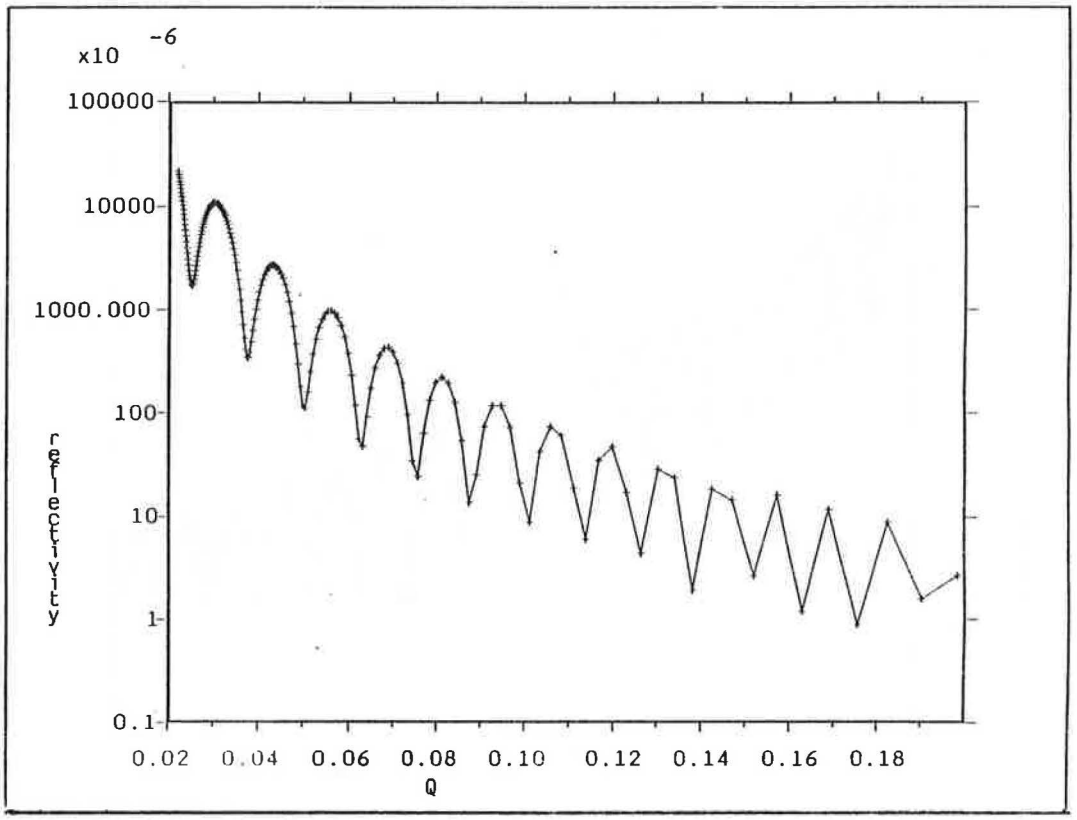


Figure 5(a)

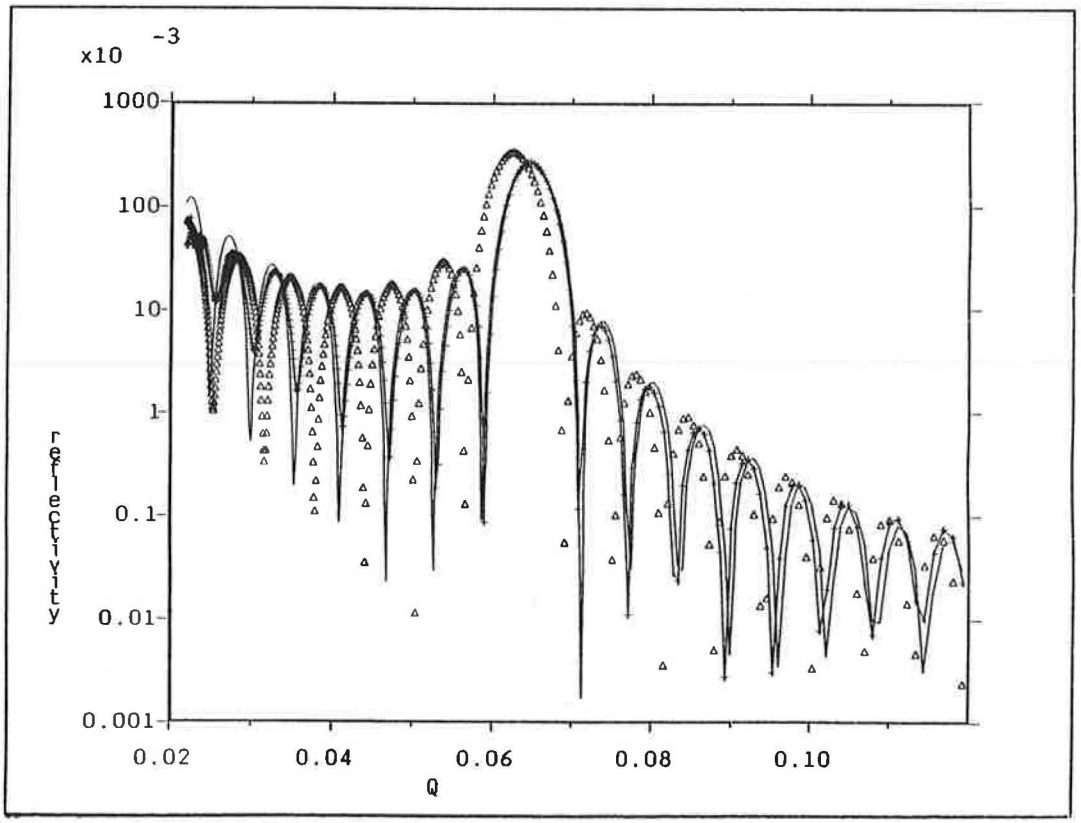
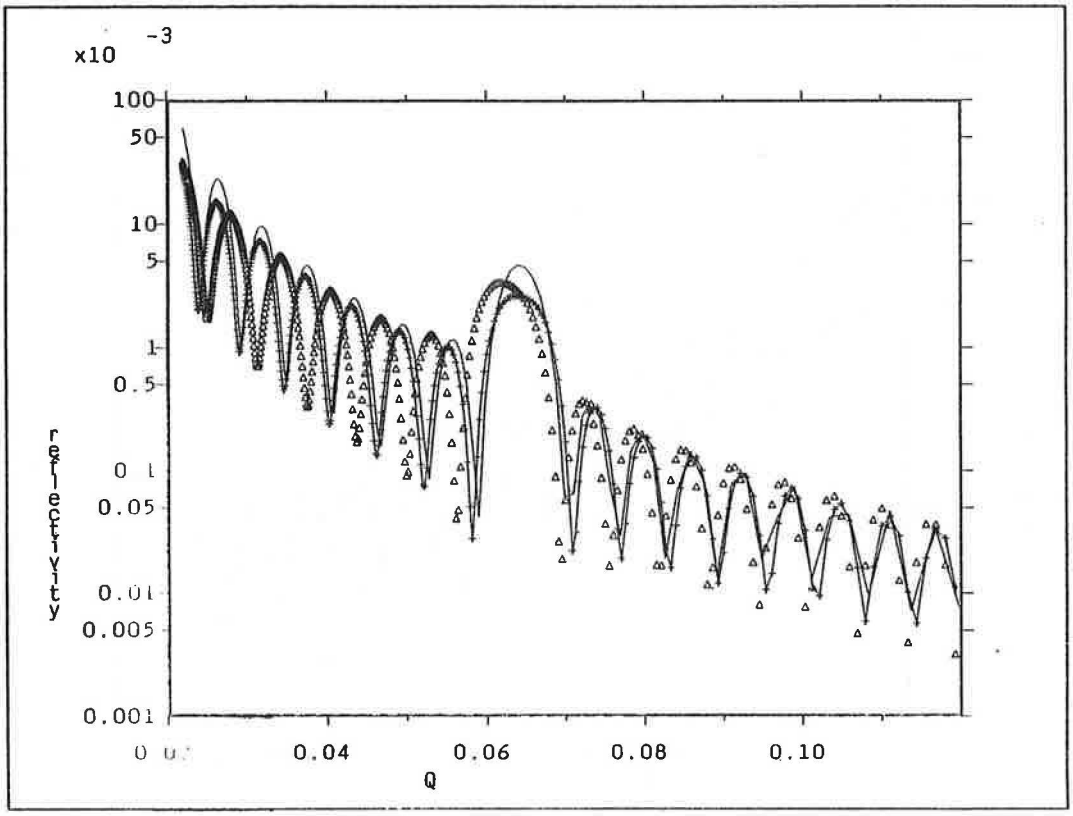


Figure 5(b)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5
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reflectivity
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0.10
Q

Figure 5(c)