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A New Instrumental Factor in Triple-Axis Spectrometry and Bragg Reflectivity Measurement

By Graham L. Tindle

Tindle Consultants Limited, Flat 7, Chesterfield House, 1B King Henry's Road, London NW3 3QP, England

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Abstract

Attention is drawn to a hitherto-neglected instrumental consideration in Bragg scattering. It requires a new sin θ factor to be included in expressions for Bragg reflectivity. The factor, which arises out of a consideration of mosaic spread is also relevant to scattering from a perfect single crystal. The scattering process is examined within a framework of neutron triple-axis spectrometry and implications of the effect for normalization of the instrumental resolution function in that connection are examined in detail.

1. Introduction

In 1958, Caglioti, Paoletti & Ricci first considered resolution effects in triple-axis neutron spectrometry. In 1963 Collins published his paper on the subject and in 1967, Cooper & Nathans proposed their Gaussian model. They described how limited resolution could be simulated by convoluting the scattering function with a Gaussian ellipsoidal distribution in four dimensions, and gave details of its form. An extension of the Cooper & Nathans formalism was subsequently employed by Samuelsen, Hutchings & Shirane (1970) and by Werner & Pynn (1971) to take account of resolution-function normalization. The following year Dorner (1972) gave a detailed physical analysis. He pointed out that for a correct intercomparison of experimental data at different wavelengths, for which in general the primary neutron beams will have different intensities and wavelength spreads, the normalization of the Cooper & Nathans (1967) Gaussian ellipsoidal function was significant. He recomputed its form.

Dorner also remarked upon what he thought to be two misprints or errors in the Cooper & Nathans (1967) paper. Firstly, vertical mosaic contributions involving monochromator and analyser Bragg scattering factors tan θ_A and tan θ_M should have been replaced by sin θ_A and sin θ_M in Cooper & Nathans (1967) equation (6), which is true. Secondly, he suggested that sin θ_A and sin θ_M factors arising from vertical mosaic integrations should be cancelled from the normalization function. The existence of such factors had not been seriously considered by previous authors and in subsequent publications (Tucciarone, Lau, Corliss, Delpalme & Hastings, 1971; Chesser & Axe, 1973) his conclusion has been endorsed. With these publications we disagree. In this study we give theoretical arguments for the recognition of sin θ factors in reflectivity measurements as reflections of a genuine physical effect and show that experimental evidence supports our claim. This is not to say that all published measurements involving the reflectivity are incorrect; in most cases a method of analysis has been used in which the recommended factor cancels out. Published expressions for resolution-function normalization factors must, however, be modified.

In the body of this paper we take the trail blazed by the published literature. Firstly, we follow the formalism of Cooper & Nathans (1967) for Bragg scattering. In § 3 we introduce Dorner's semi-classical argument (Dorner, 1972) and show where we believe it to be fallacious. In § 4 we cite confirmation of our hypothesis from published reflectivity data of Chesser & Axe (1973) and in § 5 we summarize our conclusions.

In Appendix I we parallel the content of the body of the paper with a formal development. Our principal theoretical contribution to reflectivity measurement there is embodied in (31). We give a detailed derivation in formal terms because of its general application, but also in part to allay any fears that the result arises from incorrect probability-function normalization, a possibility first suggested by Chesser in a private communication to Dorner (1972) and referred to later in his paper with Axe (Chesser & Axe, 1973).

In Appendix II we detail simple changes which can be made to expressions in the published literature (Tucciarone, Lau, Corliss, Delpalme & Hastings, 1971; Dorner, 1972; Chesser & Axe, 1973) to make them conform with our investigation.

2. Bragg scattering from a single crystal. The reflectivity

The flux $F_f(\mathbf{k}_f)$ emerging from a crystal after Bragg scattering of an incident beam of spectral distribution $F_i(\mathbf{k}_i)$ is given by equation (33) of Dorner's (1972)

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paper:

$$F_f(\mathbf{k}_f)\,d\mathbf{k}_f = N_M P_0 \exp\left\{-\frac{1}{2}\left[\left(\frac{\Delta_M+\gamma_1}{\eta_M}\right)^2+\left(\frac{2\Delta_M+\gamma_1}{\alpha_0}\right)^2 + \frac{\gamma_1^2}{\alpha_1^2}+\left(\frac{1}{\varepsilon_M^2+\beta_0^2}+\frac{1}{\beta_1^2}\right)\delta_1^2\right]\right\} F_i(\mathbf{k}_i)\,d\mathbf{k}_f, \quad (1)$$

where we follow the notation of Cooper & Nathans (1967). These authors write θ_M and k_F for the most probable Bragg scattering angle and the most probable final momentum, respectively, when these terms refer to the θ and k appropriate to scattering by mosaic elements at the peak of the mosaic distribution. In fact we have set

$$\Delta_M=(\Delta k_f/k_F)\tan\theta_M, \quad (2)$$

$$\varepsilon_M=2\eta'_M\sin\theta_M. \quad (3)$$

Subscripts M (for monochromator) have been retained here simply to allow the formulas to be used consistently in the arguments of later sections.

Divergence angles γ_1 and δ_1 in and perpendicular to the most probable scattering plane are simply related to the components of final momentum $\mathbf{k}_f$,

$$\Delta\mathbf{k}_f=\mathbf{k}_f-\mathbf{k}_F=(\Delta k_f, k_F\gamma_1, k_F\delta_1). \quad (4)$$

Parameters α_0, α_1, β_0, β_1 are the divergence angles of the collimators before and after scattering in and perpendicular to the most probable scattering plane, whilst η_M and η'_M are the in-plane (horizontal) and perpendicular (vertical) mosaic-spread widths. The normalization factor P_0 is given by

$$P_0=(2\pi)^{1/2}\left(\frac{1}{\beta_0^2}+\frac{1}{\varepsilon_M^2}\right)^{-1/2}, \quad (5)$$

and for the moment we take N_M to be an essentially energy-independent normalization factor of the form

$$N_M=(2\pi)^{-1}\frac{P_M}{\eta_M\eta'_M}. \quad (6)$$

Here we have introduced the quantity P_M that in effect functions as a reflectivity coefficient for the crystal. Explicitly it may be identified with the coefficient $\mathscr{R}(k_f)$ in (29). From (31) we find

$$P_M\equiv\mathscr{R}(k_F)=\frac{1}{2\sin\theta_M}\frac{F_{fH}(k_F,0)}{F_{iH}(k_F,0)}, \quad (7)$$

which represents it in terms of an outgoing to incoming peak-flux ratio. The most direct way of measuring it is to determine the peak-count ratio $F_{fH}(k_F,0)/F_{iH}(k_F,0)$ with no vertical collimation and a well defined θ_M, whereupon (7) applies directly. In a situation where instrumental effects are significant one may turn to § 4 and determine the reflectivity coefficient in (19). This may require a full evaluation of the integrals in (16). In any event we choose P_M to represent the intrinsic crystal reflectivity contribution to the normalization factor N_M. We justify this choice in the next section where we analyse the scattering in a manner proposed by Dorner (1972).

3. The Dorner normalization

Dorner seems to be the only person who has attempted to realize the scattering using a classical picture (Dorner, 1972). His reasoning leads to the replacement of N_M by N_{MD} (Chesser & Axe, 1973), where

$$N_{MD}=(2\pi)^{-1/2}\frac{P_{MD}}{2\eta'_M\sin\theta_M}, \quad (8)$$

and P_{MD} is his intrinsic crystal reflectivity.

Let us recall his argument.

We imagine a fan of k vectors spread in a direction normal to the most probable scattering plane. The spread angle is proportional to the incident collimation parameter β_0. Let the vectors represent neutrons incident with momentum k_1 at the most probable in-plane scattering angle for Bragg reflection. In the arrangement contemplated all emerging neutrons are counted. If the vertical scattering of our fan of neutrons is independent of the Bragg condition which determines scattering in the horizontal plane we should expect the scattering cross section to be proportional to the collimation parameter β_0 and independent of the incident Bragg angle θ_M. This is Dorner's proposition. The vertical spread of vectors should contribute to scattering in a manner independent of energy.

Let us now look at the theoretical implication. The vertical spread is represented by the δ_1 terms in (1). If we omit the final collimator, the vertical contribution to the final scattering is given by

$$F_V=P_0\int_{-\infty}^{\infty}\exp\left\{-\frac{1}{2}\left(\frac{\delta_1^2}{\varepsilon_M^2+\beta_0^2}\right)\right\}d\delta_1$$

$$=2\pi\varepsilon_M\beta_0=4\pi\beta_0\eta'_M\sin\theta_M. \quad (9)$$

The theory predicts a sin θ_M energy dependence. This we do not want, so we introduce the normalization factor N_{MD} which kills it.

In the above discussion there is an error. The scattering of the vertical fan of neutrons is not independent of the horizontal Bragg condition. For scattering to take place at all the energy of the incident neutrons is controlled. The magnitude of the momentum of the fan of incident neutrons is controlled. The fact that neutrons out of the most probable scattering plane can be scattered at all is a function of the vertical mosaic spread of the scattering crystal. The probability of scattering any such neutron depends critically on the angle the reciprocal-lattice Bragg vector of such a neutron makes with the most probable

Bragg vector lying in the most probable scattering plane. This angle for an incident wave vector of magnitude k_I is proportional to k_I and inversely proportional to $\sin\theta_M$. The geometry is displayed in Fig. 1.

The contribution to scattering we then expect is proportional to a population factor f_V, where

$$f_V = \frac{1}{(2\pi)^{1/2}\eta'_M}\exp\left\{-\frac{1}{2}\left(\frac{\delta_1-\delta_0}{2\sin\theta_M\eta'_M}\right)^2\right\},$$

where δ_0 is the angle of inclination of the incident neutron and $(\delta_1-\delta_0)/2\sin\theta_M$ is the angle of inclination of the Bragg scattering vector to the most probable scattering plane. The full contribution is proportional to

$$F'_V = \iint \exp\{-\tfrac{1}{2}(\delta_0/\beta_0)^2\} f_V \, d\delta_0 \, d\delta_1 = (2\pi)^{1/2}\beta_0 2\sin\theta_M. \quad (10)$$

With this argument the presence of the factor $\sin\theta_M$ comes as no surprise. As the Bragg angle changes so the inclination of the Bragg vector associated with an incident wave vector making a constant angle with the most probable scattering plane varies with a $\sin\theta_M$ dependence. The Dorner cancellation is not needed. The $\sin\theta_M$ dependence is real. The use of the normalization factor N_{MD} is no longer justified.

4. Experimental verification of normalization N_M

As evidence of the correctness of our formulation with N_M replacing N_{MD} we look at the paper of Chesser & Axe (1973). They give graphs of the variation of the analyser reflectivity P_{AD} with energy. (They follow Dorner's analysis.) They use a spectrometer with three crystals, a monochromator (M), a sample (S), and an analyser (A) as depicted in Fig. 2. They make measurements of reflectivity without the final collimator.

Let us repeat the calculation.

The flux of neutrons emerging from the collimator following the sample is given by a simple modification of the Cooper & Nathans expression [equation (1)],

$$F_i(\mathbf{k}_i) = F_0(k_I)N_M N_S P_0 P_1 \exp\left\{-\frac{1}{2}\left[\left(\frac{\Delta_M-2\Delta_S+\gamma_2}{\eta_M}\right)^2 + \left(\frac{2\Delta_M-2\Delta_S+\gamma_2}{\alpha_0}\right)^2 + \left(\frac{\Delta_S-\gamma_2}{\eta_S}\right)^2 + \left(\frac{\gamma_2-2\Delta_S}{\alpha_1}\right)^2 + \left(\frac{\gamma_2}{\alpha_2}\right)^2 + D_S\delta_2^2\right]\right\}, \quad (11)$$

where

$$P_1 = (2\pi)^{1/2}\left(\frac{1}{\varepsilon_M^2+\beta_0^2}+\frac{1}{\beta_1^2}+\frac{1}{\varepsilon_S^2}\right)^{-1/2} \quad (12)$$

and

$$D_S = \left[\varepsilon_S^2 + \left(\frac{1}{\varepsilon_M^2+\beta_0^2}+\frac{1}{\beta_1^2}\right)^{-1}\right]^{-1} + \frac{1}{\beta_2^2}. \quad (13)$$

Similarly, the neutron flux emergent from the analyser without a final collimator is given by

$$F_f(\mathbf{k}_f) = F_0(k_F)N_M N_S N_A P_0 P_1 P_2 \times \exp\left\{-\frac{1}{2}\left[\left(\frac{\Delta_M-2\Delta_S+2\Delta_A+\gamma_3}{\eta_M}\right)^2 + \left(\frac{2\Delta_M-2\Delta_S+2\Delta_A+\gamma_3}{\alpha_0}\right)^2 + \left(\frac{2\Delta_A-2\Delta_S+\gamma_3}{\alpha_1}\right)^2 + \left(\frac{\Delta_A-2\Delta_S+\gamma_3}{\eta_S}\right)^2 + \left(\frac{2\Delta_A+\gamma_3}{\alpha_2}\right)^2 + \left(\frac{\Delta_A+\gamma_3}{\eta_A}\right)^2 + (\varepsilon_A^2+D_S^{-1})^{-1}\delta_3^2\right]\right\}, \quad (14)$$

where

$$P_2 = (2\pi)^{1/2}(D_S+\varepsilon_A^{-2})^{-1/2}. \quad (15)$$

We are interested in the ratio Λ of scattering amplitudes before and after the analyser, where

$$\Lambda = \int F_f(\mathbf{k}_f)\, d\mathbf{k}_f \Big/ \int F_i(\mathbf{k}_i)\, d\mathbf{k}_i. \quad (16)$$

In general this will be a complicated function of the most probable energy ω_F. The integrals can be separated into horizontal contributions dependent on Δk

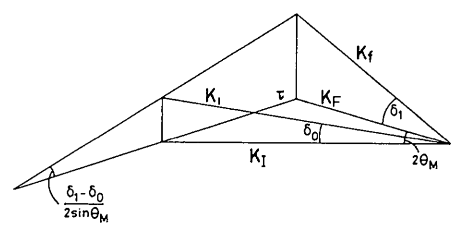

Fig. 1. Geometry of the $\sin\theta_M$ factor.

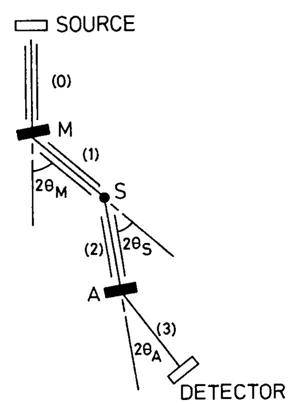

Fig. 2. The spectrometer configuration.

and γ_2 or γ_3 and a vertical contribution dependent upon δ_2 or δ_3, such that

$$\Lambda = N_A \Lambda_H \Lambda_V. \tag{17}$$

If we make the reasonable approximation $\Delta_M = \Delta_S = \Delta_A$ one can show by direct computation that the horizontal component of the ratio Λ_H becomes independent of energy. The vertical component is then given by

$$\Lambda_V = (2\pi)^{1/2} \varepsilon_A. \tag{18}$$

The whole ratio takes the form

$$\Lambda = (2\pi)^{1/2} N_A \Lambda_H \varepsilon_A, \tag{19}$$

which has the simple sin θ_A energy dependence predicted in a simple way by (9) and (10). Now with the Dorner normalization N_{AD} this sin θ_A dependence is explicitly eliminated and theoretically we should expect it to reappear in the experimentally measured reflectivity parameter P_{AD}. Whereas Dorner would expect the experimentally measured reflectivity P_{AD} to be only a slowly varying function of energy, reflecting an intrinsic property of the crystal material, we predict that in addition it should vary in proportion to sin θ_A or inversely in proportion to k_F or $\omega_F^{1/2}$. A glance at the published reflectivity curves of Chesser & Axe (1973) reveals that the variation with energy is almost wholly described (Fig. 3) by an $\omega_F^{-1/2}$ variation. Indeed, the variation in 002 scattering is, within experimental error, totally of this form. We suggest that the variation should be regarded as a consequence of the process of measurement and not indicative of any intrinsic crystal property. This view indicates the choice of an essentially constant function for the normalization function N_A. We propose (6) for its form and choose P_A to represent the intrinsic reflectivity. Then we find experimentally that P_A being proportional to $\omega_F^{1/2} P_{AD}$ is almost a constant function of energy.

5. Conclusions

We have demonstrated that Dorner's approach to the problem of Bragg scattering proves useful when one wishes to analyse the energy dependence of the Cooper & Nathans resolution function in triple-axis spectrometry. Our application of these ideas leads us to the opinion that, contrary to what Dorner proposed in his original paper (Dorner, 1972), the original formulation of Cooper & Nathans (1967) is not in error. When one takes into account a realistic picture of a crystal with mosaic spread the appearance of a sin θ factor in the measured reflectivity becomes a natural physical feature. Furthermore the feature is clearly evident in the reflectivity curves published in 1973 by Chesser & Axe. Our researches suggest that the resolution function specified by Chesser & Axe be redefined, N_M and N_A being given by (6) rather than (8). In addition, this modification should be used in any application to inelastic neutron scattering where P_M and P_A may be assumed constant.

We hope that our discussions give a clearer picture of the features of Bragg scattering relevant to the normalization of the resolution function and that it will prove useful in future applications to spectrometer measurements of neutron scattering where Bragg reflectivity is of significance.

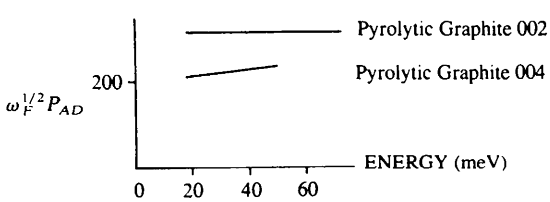

Fig. 3 Energy variation of $\omega_F^{1/2} P_{AD}$.

I am particularly grateful to Drs R. D. Lowde and M. T. Hutchings at UKAEA Harwell for suggested improvements to my original manuscript and their interest. I have received from them notice of other reflectivity curves, all of which exhibit the $\omega^{-1/2}$ energy dependence.

APPENDIX 1

A formal derivation of the reflectivity factor

Let the mosaic spread of a crystal be defined by a separable population-density distribution function P of vertical and horizontal angular displacements ζ, ξ of Bragg lattice vector from the most probable vector $\boldsymbol{\tau}$ such that in coordinate axes relative to $\boldsymbol{\tau}$ we have

$$\Delta \boldsymbol{\tau} = (\Delta \tau, \tau\xi, \tau\zeta). \tag{20}$$

The sharply peaked separable population distribution must be normalized with respect to these angular displacements. We have

$$P(\xi, \zeta) = P_H(\xi) P_V(\zeta), \tag{21}$$

where

$$\int_{-\infty}^{\infty} P_V(\zeta)\, d\zeta = \int_{-\infty}^{\infty} P_H(\xi)\, d\xi = 1. \tag{22}$$

We now consider Bragg scattering with flux $F_i(\mathbf{k}_i)\, d\mathbf{k}_i$ incident upon the crystal and emergent flux $F_f(\mathbf{k}_f)\, d\mathbf{k}_f$, where, following the notation of Cooper & Nathans (1967), we have

$$\mathbf{k}_i = \mathbf{k}_I + \Delta \mathbf{k}_i, \quad \mathbf{k}_f = \mathbf{k}_F + \Delta \mathbf{k}_f, \tag{23}$$

$$\Delta \mathbf{k}_i = (\Delta k_i, k_I \gamma_0, k_I \delta_0), \quad \Delta \mathbf{k}_f = (\Delta k_f, k_F \gamma_1, k_F \delta_1), \tag{24}$$

in coordinate systems relative to vectors $\mathbf{k}_I$ and $\mathbf{k}_F$, respectively.

Now for Bragg scattering we have

$$k_i = k_f,\ k_I = k_F = k. \tag{25}$$

Since $\Delta\boldsymbol{\tau} = \Delta\mathbf{k}_f - \Delta\mathbf{k}_i$ we find

$$\Delta\boldsymbol{\tau} = \left[0,\ \tau\left(\frac{\gamma_0+\gamma_2}{2}\right),\ \tau\left(\frac{\delta_1-\delta_0}{2\sin\theta}\right)\right], \tag{26}$$

where

$$2k\sin\theta = \tau. \tag{27}$$

Differentiation of the constraint (27), keeping τ constant, leads to the relation

$$\gamma_0 = \gamma_1 + 2\frac{\Delta k_f}{k}\tan\theta. \tag{28}$$

This shows us (Cooper & Nathans, 1967) that the angle of incidence γ_0 is uniquely constrained to be a function of the emergent angle γ_1 and final momentum k_f.

We are now in a position to record an expression for the emergent flux,

$$F_f(\mathbf{k}_f)\,\mathrm{d}\mathbf{k}_f = \left[\int_{-\infty}^{\infty} F_i(\mathbf{k}_i) \times P\left(\frac{\gamma_0+\gamma_1}{2}, \frac{\delta_1-\delta_0}{2\sin\theta}\right)\mathrm{d}\delta_0 \mathcal{R}(k_f)\right]\mathrm{d}\mathbf{k}_f. \tag{29}$$

It represents the emergent flux for an incident beam with γ_0, restricted by (28), arising from contributions with a full range of incident vertical divergence angles δ_0. Our energy-dependent physical reflectivity is $\mathcal{R}(k_f)$.

We shall now elucidate the effect of vertical components of the beam. We replace $\mathrm{d}\mathbf{k}_f$ by $k_f^2\,\mathrm{d}k_f\,\mathrm{d}\gamma_1\,\mathrm{d}\delta_1$ and integrate over δ_1 to obtain the flux as a function of horizontal divergence angle and final momentum,

$$F_{fH}(k_f, \gamma_1)\,\mathrm{d}k_f\,\mathrm{d}\gamma_1 = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} F_i(\mathbf{k}_i) \times P\left(\frac{\gamma_1+\gamma_0}{2}, \frac{\delta_1-\delta_0}{2\sin\theta}\right)\mathrm{d}\delta_0\,\mathrm{d}\delta_1 \times \mathcal{R}(k_f)\,\mathrm{d}k_f\,\mathrm{d}\gamma_1. \tag{30}$$

We change from variable δ_1 to x, where $x = (\delta_1-\delta_0)/2\sin\theta$, and integrate to obtain the relation

$$F_{fH}(k_f, \gamma_1)\,\mathrm{d}k_f\,\mathrm{d}\gamma_1 = 2\sin\theta F_{iH}(k_i, \gamma_0) \times P_H\left(\frac{\gamma_1+\gamma_0}{2}\right)\mathcal{R}(k_f)\,\mathrm{d}k_f\,\mathrm{d}\gamma_1, \tag{31}$$

where

$$F_{iH}(k_i, \gamma_0) = \textstyle\int_{-\infty}^{\infty} F_i(\mathbf{k}_i)\,\mathrm{d}\delta_0. \tag{32}$$

The vertical mosaic effects have been such as to introduce an overall factor $2\sin\theta$ into our expression for the emergent horizontal flux. The factor $2\sin\theta$ is universal. The effect of horizontal mosaic is not, but, in the Gaussian approximation considered in the body of this paper, in reflectivity measurement horizontal effects cancel out. The true reflectivity function $\mathcal{R}(k_f)$ becomes multiplied by the instrumental factor $2\sin\theta$ which is proportional to $1/k_F$ or $\omega_F^{-1/2}$. In the past it is this anomalous reflectivity product which has been measured.

It was important in deriving the factor $2\sin\theta$ that we had no vertical collimation between crystal and detector. Such an arrangement would introduce another function $C(\delta_1)$ into our integrands and after transformation the x integral could not then be evaluated. The more complex effect of the vertical component could however be then computed in a Gaussian approximation.

APPENDIX II

Changes to published literature

Our observations suggest that in the papers by Dorner (1972) and Chesser & Axe (1973), in all occurrences, P_M and P_A should be replaced as follows:

$$P_M \rightarrow \frac{2P_M\sin\theta_M}{(2\pi)^{1/2}\eta_M},\quad P_A \rightarrow \frac{2P_A\sin\theta_A}{(2\pi)^{1/2}\eta_A}. \tag{33}$$

For consistency, apart from corrections detailed in the Appendix of Chesser & Axe (1973), in Tucciarone, Lau, Corliss, Delpalme & Hastings (1971) the following replacements should be effected:

$$P_M \rightarrow \frac{2P_M\sin\theta_M}{2\pi\eta_M\eta'_M},\quad P_A \rightarrow \frac{2P_A\sin\theta_A}{2\pi\eta_A\eta'_A}. \tag{34}$$

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