two-parameter theory of Sec. III A. In comparison with a previously calculated Compton profile using Kunz wave functions,[18] the present tight-binding calculation is in much better agreement with experiment. The effect of overlaps on the scattering factors is presently being investigated.

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Lattice Dynamics above Structural Phase Transitions: $SrTiO_3$

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The temperature dependence of the soft mode ω_0, the central peak $S(\vec{q}, \omega=0)$, and the electron-paramagnetic-resonance (EPR) linewidth ΔH above a continuous structural phase transition driven by a soft R-corner mode is calculated. In the noncritical region the important self-energy terms leading to a central peak at $\vec{q}=\vec{q}_R$ are shown to be the chain diagrams. This is due to a particular feature of the lowest transverse phonon branch giving rise to a two-phonon density of states $\rho_0(\vec{q}, \omega)$ which is nonzero for $\vec{q}\cong\vec{q}_R, \omega\cong 0$. The exponents of $T-T_c$ found for ω_0, $S(\vec{q}_R, \omega=0)$, and ΔH are, respectively, $+\frac{1}{2}$, -2, and $-\frac{1}{2}$, the first being the same as that obtained in the mean-field theory of Pytte and Feder, and the second agreeing with a result of Cowley.

I. INTRODUCTION

In this paper a unified lattice-dynamical description of continuous structural phase transitions driven by a soft mode at the R corner $\vec{q}_R=(\pi/a)(1, 1, 1)$ of the Brillouin zone is proposed. The present situation of the problem is as follows.

(a) In $SrTiO_3$ ($T_c=105$ K) the temperature dependence of the soft mode was explained by Pytte and Feder[1] as a Hartree renormalization effect. Unfortunately, their harmonic frequency turned out to be imaginary, as in the work of Cowley.[2]

(b) The connection of a phase transition with a driving soft mode was recently formalized by showing that the isothermal order-parameter susceptibility has a pole which moves to the origin as $T\to T_c$.[3]

(c) Inelastic neutron and Raman scattering have recently revealed a central peak $S(\vec{q}, \omega=0)$ near T_c in[4] $SrTiO_3$ and in other crystals.[5,6] An explanation in terms of a diffusive ($\vec{q}=0$) Landau–Placzek peak was given by Feder[7] which, however, causes difficulties in the case $\vec{q}=\vec{q}_R$. This case may be understood, in principle, in terms of a bubble diagram where one phonon line is replaced by a ladder insertion.[8]

(d) A phenomenological description of a central peak in terms of a particular frequency-dependent damping function $\Gamma(\omega)$ [Eq. (27) below] has been proposed by Shirane and Axe[6] and by Schwabl.[9] This function [but with $\Gamma(\infty)=0$ as in Eq. (27)] has recently been shown by Schneider[10] to follow from

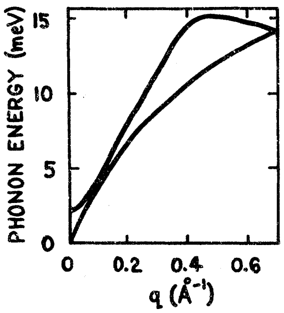

FIG. 1. Dispersion curve $\omega_{\vec{q}}$ of the lowest transverse branch of $SrTiO_3$ at $T^* = 120$ K in the [1 1 1] direction plotted in the reduced-zone representation $q < \frac{1}{2} q_R$, taken from Fig. 3 of Ref. 12.

a continued-fraction representation. Making use of the connection mentioned in (b) Schneider also confirmed Feder's result.[7]

(e) The EPR linewidth ΔH has recently been measured by Müller *et al.*[11] as a function of $\epsilon = (T - T_c)/T_c$. They produced a critical exponent $\nu = \frac{2}{3}$ which Schwabl[9] related to critical order-parameter fluctuations.

In terms of lattice dynamics, $\Gamma(\omega)$ is related to the self-energy of the soft mode. In order to produce the denominator of the phenomenological $\Gamma(\omega)$ of Eq. (27) below, it is sufficient to sum up the infinite set of chain diagrams. The reason for the importance of the chain diagrams lies in a remarkable property of the dispersion curve $\omega_{\vec{q}}$ of the relevant mode. Since below T_c the R corner is equivalent with the zone center, $\vec{q}_R \sim 0$, a reduced-zone representation with $q < \frac{1}{2} q_R$ in the [111] direction is appropriate. In this representation, the measured dispersion curve $\omega_{\vec{q}}$ for $SrTiO_3$ at a fixed temperature[12] $T^* > T_c$ consists of two almost touching branches as exhibited in Fig. 1. The situation is similar for[13] $KMnF_3$ and, less pronounced, for $LaAlO_3$.[14] As a consequence of this feature the two-phonon density of states (its particular form is explained below)

$$\rho_0(\vec{q}, \omega) = (1/N) \sum_{\vec{k}}{}' f_0(\vec{k}, \vec{q}) \delta(\omega + \omega_{\vec{k}} - \omega_{\vec{q}-\vec{k}}) \quad (1)$$

is nonzero at the relevant values $\omega \cong 0$ and $\vec{q} \cong \vec{q}_R$. This fact gives rise to an enhancement of diagram parts consisting of two-phonon bubbles with frequency $\sim (\omega_{\vec{q}-\vec{k}} - \omega_{\vec{k}})$, and consequently leads to a dominance of the chain diagrams.

II. SELF-ENERGY

The general expression of the self-energy above T_c can be written[15]

$$\Pi = \Pi_H + \Pi_3 + \cdots, \quad (2)$$

where

$$\Pi_H(\vec{q}) = \sum_{\vec{k}, \nu} \gamma_4(\vec{q}, \vec{k}, -\vec{k}) D(\vec{k}, \nu) \quad (3)$$

is the Hartree term studied by Pytte and Feder[1] and

$$\Pi_3(\vec{q}, \omega) = \sum_{\vec{k}, \nu} \gamma_3(\vec{q}, \vec{k}) D(\vec{k}\nu) \times D(\vec{q} - \vec{k}, \omega - \nu) \Gamma_3(\vec{q}, \omega; \vec{k}, \nu) . \quad (4)$$

Similar but increasingly complicated expressions hold for the higher-order terms. Here γ_n and Γ_n are the bare and renormalized n-point vertex functions, respectively; D is the renormalized propagator; and $\nu = \pm (2\pi i/\beta) \times$ integer. The dominance of the two-phonon bubbles at $\vec{q} \cong \vec{q}_R$, $\omega \cong 0$ owing to the density of states (1) implies that the higher-order terms in (2) are negligible, and that Γ_3 in (4) is dominated by the chain diagrams

$$\Gamma_3(\vec{q}, \omega; \vec{k}_1) = \gamma_3(\vec{q}, \vec{k}_1) + \sum_{N=2}^{\infty} \sum_{\vec{k}_2, \nu_2 \cdots \vec{k}_N, \nu_N} \left\{ \prod_{S=2}^{N} \gamma_4(\vec{q}, \vec{k}_{S-1}, \vec{k}_S) \times D(\vec{k}_S, \nu_S) D(\vec{q} - \vec{k}_S, \omega - \nu_S) \right\} \gamma_3(\vec{q}, \vec{k}_N) . \quad (5)$$

For the explicit calculation we choose 3- and 4-phonon interactions with factorizing vertex functions γ_n,

$$H_n = \frac{4\lambda_n}{n!} N^{1-n/2} \sum_{\vec{q}_1 \cdots \vec{q}_n} (\gamma^{(n)}_{\vec{q}_1} \cdots \gamma^{(n)}_{\vec{q}_n})^{1/2} \times Q_{\vec{q}_1} \cdots Q_{\vec{q}_n} \Delta\left(\sum_{S=1}^{n} \vec{q}_S \right) . \quad (6)$$

As shown in the Appendix, this form of H_4 follows, under particular conditions, from the quartic interaction used in Ref. 1. With (6), the Hartree term of Pytte and Feder simply is

$$\Pi_H(\vec{q}) = \lambda_4 \gamma^{(4)}_{\vec{q}} a , \quad (7)$$

where

$$a(T) = \frac{1}{N} \sum_{\vec{k}} \gamma^{(4)}_{\vec{k}} \coth(\tfrac{1}{2} \beta \omega_{\vec{k}}) \quad (8)$$

is a positive monotonically increasing function of T.

From (4), (5) and with the constant-lifetime approximation[16]

$$D(\vec{q}, \omega) = -\int_0^{\beta} d\tau\, e^{\omega\tau} \langle \mathcal{T}(Q_{\vec{q}}(-i\tau) Q_{-\vec{q}}(0)) \rangle = \omega_{\vec{q}} [(\omega + \tfrac{1}{2} i \Gamma_0)^2 - \omega_{\vec{q}}^2]^{-1} , \quad (9)$$

the self-energy of the chain diagrams is found to be

$$\Pi_3(\vec{q}, \omega) = 4\lambda_3^2 \gamma^{(3)}_{\vec{q}} G(\vec{q}, \omega) . \quad (10)$$

Here

$$G = G_0 (1 - \lambda_4 G_0)^{-1} \quad (11)$$

and

$$G_0(\vec{q}, \omega) = \int_{-\infty}^{+\infty} d\omega'\, \rho(\vec{q}, \omega') \frac{\omega'^2}{(\omega + i\Gamma_0)^2 - \omega'^2} \quad (12)$$

are, respectively, the renormalized and bare propagators of the collective coordinate

$$\epsilon_{\vec q} = N^{-1/2} \sum_{\vec k}{}' (\gamma^{(4)}_{\vec k}\gamma^{(4)}_{\vec q-\vec k})^{1/2} Q_{\vec k} Q_{\vec q-\vec k} \qquad (13)$$

defined as D in Eq. (9). Thus, the pole of G as given by

$$G_0(\vec q, \omega) = \lambda_4^{-1} \qquad (14)$$

represents a collective mode of the interacting phonons. With $\gamma^{(4)}_{\vec q} = \omega_{\vec q}$ (see the Appendix), $\epsilon_{\vec q}$ in Eq. (13) is recognized as the energy density of the phonons. Hence G is the energy-density correlation function and the collective mode describes energy propagation. Both the structure of such a collective-mode propagator in an anharmonic linear chain[17] and its possible effect through Eq. (10) on the sound propagation in superfluid helium have been studied before.[18] As we shall see, however, it is not this collective mode which is important for the existence of a central peak in $S(\vec q, \omega)$, but the behavior for $\omega \cong 0$ of the density of states in Eq. (12). It consists of two terms,

$$\rho = \rho_0 + \rho_1 , \qquad (15)$$

where ρ_0 is given by (1) and

$$f_0(\vec k, \vec q) = \gamma^{(4)}_{\vec k}\gamma^{(4)}_{\vec q-\vec k} \frac{n(\omega_{\vec k}) - n(\omega_{\vec q-\vec k})}{\omega_{\vec q-\vec k} - \omega_{\vec k}} \qquad (16)$$

is a positive monontonically increasing function of T (Ref. 16) and $n(\omega) = (e^{\beta\omega} - 1)^{-1}$. ρ_1 has the same form (1) but with

$$f_1(\vec k, \vec q) = \gamma^{(4)}_{\vec k}\gamma^{(4)}_{\vec q-\vec k} \frac{n(\omega_{\vec k}) + n(\omega_{\vec q-\vec k}) + 1}{\omega_{\vec q-\vec k} + \omega_{\vec k}} , \qquad (17)$$

which is again positive and monotonically increasing in T,[16] and with the argument $\omega - \omega_{\vec k} - \omega_{\vec q-\vec k}$ of the δ function. It is obvious from this that $\rho_1 = 0$ for $\omega < \omega_{\vec q_R}$ and therefore, as we shall see, does not contribute to the central peak of $S(\vec q, \omega)$. For the following discussion it is still convenient to consider the complete form (15). ρ is nonzero in a finite interval,

$$\rho(\vec q, \omega) \neq 0 , \quad \Omega^-_{\vec q} < \omega < \Omega^+_{\vec q} , \qquad (18)$$

the extremities $\Omega^+_{\vec q}$ and $\Omega^-_{\vec q}$ being defined, respectively, as maximum and minimum of $\omega_{\vec k} \pm \omega_{\vec q-\vec k}$ with respect to $\vec k$. In the neighborhoods of $\Omega^+_{\vec q}$ and $\Omega^-_{\vec q}$, $\rho(\vec q, \omega)$ therefore behaves as $(\Omega^+_{\vec q} - \omega)^{1/2}$ and $(\omega - \Omega^-_{\vec q})^{1/2}$, respectively. A similar cusp-shaped behavior occurs at any other extremum of $\omega_{\vec q-\vec k} \pm \omega_{\vec k}$. For $\vec q = \vec q_R$, the extremity $\Omega^-_{\vec q_R} > 0$ if the two branches in Fig. 1 do not touch and $\Omega^-_{\vec q_R} < 0$ if they cross. For $\vec q \neq \vec q_R$, the two branches in Fig. 1 are shifted by $\vec q_R - \vec q$ relative to each other so that the chances for crossing are increased and one has always $\Omega^-_{\vec q} < 0$ for sufficiently large $|\vec q_R - \vec q|$. This is important in what follows. It is obvious from this discussion that $|\Omega^-_{\vec q}| \ll \Omega^+_{\vec q}$.

III. DENSITY CORRELATION FUNCTION

The density correlation function $S(\vec q, \omega)$ is related, through the fluctuation-dissipation theorem, to the phonon propagator $D(\vec q, \omega)$ by

$$S(\vec q, \omega) = \pi^{-1} n(-\omega) \operatorname{Im} D(\vec q, \omega) . \qquad (19)$$

In terms of the self-energy (2), the propagator has the form[16]

$$D(\vec q, \omega) = \omega_{\vec q}[\omega^2 - \omega^2_{\vec q} - \omega_{\vec q}\Pi(\vec q, \omega)]^{-1} . \qquad (20)$$

For frequencies such that $\beta\omega \ll 1$, insertion of (7) and (10) leads to

$$S(\vec q, \omega) \propto \omega^{-1} \operatorname{Im}[\omega^2_{\vec q} + \lambda_4 \omega_{\vec q}\gamma^{(4)}_{\vec q} a + 4\lambda_3^2 \omega_{\vec q}\gamma^{(3)}_{\vec q} G(\vec q, \omega) - \omega^2]^{-1} . \qquad (21)$$

Now the multiple-fraction development of Schneider[10] corresponds to an expansion of $G_0(\vec q, \omega)$ in powers of ω. We write

$$-G_0(\vec q, \omega) = A_0 + i\omega A_1 + \omega^2 R(\vec q, \omega) , \qquad (22)$$

where

$$A_n(\vec q, T) = (2\Gamma_0)^n \int_{-\infty}^{+\infty} d\omega\, \rho(\vec q, \omega)\omega^2(\Gamma_0^2 + \omega^2)^{-n-1} \qquad (23)$$

are positive monotonically increasing functions of T. According to (12) and (18), G_0 is analytic in the complex ω plane except for a cut at $\Omega^-_{\vec q} < \operatorname{Re}\omega < \Omega^+_{\vec q}$, $\operatorname{Im}\omega = -\Gamma_0$. Since the remainder R is finite for $\omega = 0$,

$$R(\vec q, 0) = \int_{-\infty}^{+\infty} d\omega\, \rho(\vec q, \omega)\omega^2 (\omega^2 - 3\Gamma_0^2)(\Gamma_0^2 + \omega^2)^{-3} , \qquad (24)$$

it has the same analyticity property as G_0.

For sufficiently low frequencies such that the remainder R can be neglected in (22), we find, from (21) and (11),

$$S(\vec q, \omega) \propto \omega^{-1} \operatorname{Im}[\omega_0^2 - \omega^2 - i\omega\Gamma(\omega)]^{-1} , \qquad (25)$$

where

$$\omega_0^2(\vec q, T) = \omega^2_{\vec q} + \lambda_4\omega_{\vec q}\gamma^{(4)}_{\vec q}[a - gA_0/(\lambda_4^{-1} + A_0)] ,$$
$$g(\vec q) = 4(\lambda_3/\lambda_4)^2 \gamma^{(3)}_{\vec q}/\gamma^{(4)}_{\vec q} , \qquad (26)$$

and $\Gamma(\omega)$ has the form of Refs. 6 and 9 but, as in Ref. 10, without the additive constant

$$\Gamma(\omega) = \delta^2/(\gamma - i\omega) , \qquad (27)$$

with

$$\gamma(\vec q, T) = -(\lambda_4^{-1} + A_0)/A_1 ,$$
$$\delta^2(\vec q, T) = -g\omega_{\vec q}\gamma^{(4)}_{\vec q}/(\lambda_4^{-1} + A_0) . \qquad (28)$$

Here the only important sign is that $\gamma\delta^2 > 0$, which ensures causality.

For $\omega \gg \Omega^+_{\vec q}$ we have from (12) and (18)

$$G_0(\vec q, \omega) = \lambda_4^{-1}\Omega^2_\infty(\vec q)(\omega + i\Gamma_0)^{-2} , \qquad (29)$$

and from (11)

$$G(\vec{q}, \omega) = \lambda_4^{-1}\Omega_\infty^2(\vec{q})\,[(\omega + i\Gamma_0)^2 - \Omega_\infty^2(\vec{q})]^{-1} . \tag{30}$$

$\lambda_4\Omega_\infty^2$ is given by the second moment of ρ, and hence $\Omega_\infty \lesssim \Omega_{\vec{q}}^+$, unless $\lambda_4 A_0$ (taken in the limit $\Gamma_0 \to 0$) is large compared to unity. Therefore, $\omega \gg \Omega_\infty$ in Eq. (30).

Inserting (30) into (21) where the factor ω^{-1} is to be replaced by 1, since $\beta\omega \gg 1$, we find

$$S(\vec{q}, \omega \to \infty) \propto (-8\Gamma_0\lambda_4 g\omega_{\vec{q}}\gamma_{\vec{q}}^{(4)}\Omega_\infty^2)\omega^{-7} . \tag{31}$$

Hence the fourth moment of $S(\vec{q}, \omega)$, which determines the frequency ω_A of Schneider,[10] exists. (Note that in the notation of this paper $\omega_A^2 = \omega_0^2 + \delta^2$.) In the limit $\Gamma_0 \to 0$, all higher moments exist since

$$\mathrm{Im}G_0 = -(\tfrac{1}{2}\pi)\omega[\rho(\vec{q}, \omega) + \rho(\vec{q}, -\omega)] \tag{32}$$

vanishes for $\omega > \Omega_{\vec{q}}^+$. Then Eqs. (23) and (24) become

$$A_0 = \int_{-\infty}^{+\infty} d\omega\, \rho(\vec{q}, \omega) = (1/N)\sum_{\vec{k}} [f_0(\vec{k}, \vec{q}) + f_1(\vec{k}, \vec{q})] ,$$
$$A_1 = 2\int_{-\infty}^{+\infty} dx\, \rho(\vec{q}, \Gamma_0 x)x^2(1 + x^2)^{-2}\Big|_{\Gamma_0 \to 0} = \pi\rho_0(\vec{q}, 0) \tag{33}$$

and

$$R(\vec{q}, 0) = -\Gamma_0 \int_{-\infty}^{+\infty} dx\, \rho(\vec{q}, \Gamma_0 x)\frac{d}{dx}\frac{x^2}{1 + x^2}\Bigg|_{\Gamma_0 \to 0}$$
$$= \int_0^\infty \frac{d\omega}{\omega}\frac{\partial}{\partial\omega}[\rho(\vec{q}, \omega) + \rho(\vec{q}, -\omega)] , \tag{34}$$

which is finite.

The high-frequency limit of the propagator is, according to (20), (7), (10), and (30),

$$D(\vec{q}, \omega \to \infty) = \omega_{\vec{q}}(\omega^2 - \omega_\infty^2)^{-1} , \tag{35}$$

where

$$\omega_\infty^2(\vec{q}, T) = \omega_{\vec{q}}^2 + \omega_{\vec{q}}\lambda_4\gamma_{\vec{q}}^{(4)} a . \tag{36}$$

ω_∞ is the renormalized frequency of Pytte and Feder,[1] who determine the transition temperature T_c in mean-field approximation[19] from the condition $\omega_\infty(\vec{q}_R, T_c) = 0$. As mentioned in Sec. I, this leads, unfortunately, to an imaginary harmonic frequency, $\omega_{\vec{q}_R}^2 < 0$.[16] In Sec. IV we will see that in our theory T_c is determined instead by ω_0.

Comparison of Eqs. (35) and (9) shows that the limit $\omega \to \infty$ is not consistently described in our theory. We expect Eq. (9) to be a good approximation in the domain of the one-phonon pole, $|\omega| \lesssim \max\omega_{\vec{q}}$. This implies that our expression for the density of states $\rho(\vec{q}, \omega)$ also is expected to be valid in this domain, and hence that Eqs. (29) to (31) should not be taken too seriously. This does not affect, however, the low-frequency behavior which is the main interest of this paper. For small ω, comparison of (25) with (19) shows that

$$D(\vec{q}, \omega) \cong \omega_{\vec{q}}[\omega^2 + i\omega\Gamma(0) - \omega_0^2]^{-1} , \tag{37}$$

and we obtain self-consistency by requiring that Eqs. (37) and (9) coincide in the vicinity of the temperature T^* of zero renormalization.[16] This implies that, for $T \cong T^*$,

$$\Gamma_0 \cong \Gamma(0) = \delta^2/\gamma , \quad \tfrac{1}{4}\Gamma_0^2 \cong \omega_0^2 - \omega_{\vec{q}}^2 . \tag{38}$$

These conditions will be examined in Sec. V.

IV. SOFT MODE

Since the soft mode is the low-frequency pole of $S(\vec{q}_R, \omega)$, we conclude from (25) that the transition temperature is determined as the first zero of

$$\omega_0(\vec{q}_R, T_c) = 0 \tag{39}$$

coming from high temperatures. In order to investigate the temperature dependence of ω_0, Eq. (26), we need more explicit expressions for the positive monotonically increasing functions $a(T)$ and $A_0(\vec{q}_R, T)$. From (8) we conclude that

$$\varphi_1(T) \equiv \frac{a(T)}{a(0)} = \begin{cases} 1 + O(T^4) \\ (T/\theta_1)[1 + O(T^{-2})] \end{cases} . \tag{40}$$

Taking the limit $\Gamma_0 \to 0$ of A_0 we find from (33), (16), and (17)

$$\varphi_2(T) \equiv \lambda_4 A_0(\vec{q}_R, T) = \begin{cases} \varphi_2(0) + O(T^4) \\ (T/\theta_2)[1 + O(T^{-2})] \end{cases} . \tag{41}$$

Introducing the abbreviations

$$\lambda_4(\gamma_{\vec{q}_R}^{(4)}/\omega_{\vec{q}_R})\, a(0) = c_1 , \tag{42}$$

$$g(\vec{q}_R)/a(0) = c_2/c_1 , \tag{43}$$

Eq. (26) becomes, for $\vec{q} = \vec{q}_R$,

$$\omega_0^2(\vec{q}_R, T)/\omega_{\vec{q}_R}^2 = 1 + c_1\varphi_1(T) - c_2\varphi_2(T)[1 + \varphi_2(T)]^{-1} . \tag{44}$$

We require this expression to be positive in the limit $T \to \infty$.[16] This implies that $c_1 > 0$ and hence that $c_2 > 0$ and $\lambda_4 > 0$. From the latter condition we conclude that $\varphi_2(0)$ and θ_2 are positive, as is θ_1.

In order for Eq. (44) to have zeros it is necessary that the minimum at $T = 0$ of $1 + c_1\varphi_1$ must be smaller than the asymptote at $T \to \infty$ of $c_2\varphi_2/(1 + \varphi_2)$, or that

$$1 + c_1 < c_2 . \tag{45}$$

For $c_2/(1 + c_1) \gg 1$, the highest zero is $T_c \gg \theta_2$, and we find from (44), (40), and (41)

$$T_c/\theta_1 = (c_2 - 1)/c_1 > 1 . \tag{46}$$

For $T > T_c$, Eq. (44) is a monotonically increasing function of T. Hence there exists a temperature $T^* > T_c$ such that

$$\omega_0(\vec{q}_R, T^*) = \omega_{\vec{q}_R} . \tag{47}$$

For $c_2/(1 + c_1) \gg 1$, we find

$$T^*/\theta_1 = c_2/c_1 \gg 1 \tag{48}$$

or

$$\epsilon^* \equiv (T^* - T_c)/T_c = (c_2 - 1)^{-1} . \quad (49)$$

The explicit temperature dependence of (44) is easily found to be

$$\omega_0^2(\vec{q}_R, T)/\omega_{\vec{q}_R}^2 = \epsilon/\epsilon^* = (T - T_c)/(T^* - T_c) , \quad (50)$$

valid for $c_2/(1+c_1) \gg 1$, $T_c \gg \theta_2$. The temperature dependence of Eq. (50) is identical with that of the mean-field approximation[19] of Ref. 1. On the other hand, we have, from Eqs. (36), (40), (48), and (49),

$$\omega_\infty^2(\vec{q}_R, T)/\omega_{\vec{q}_R}^2 = 1 + c_1\varphi_1(T) \cong 1 + (1 + 1/\epsilon^*)T/T^* . \quad (51)$$

To get a feeling for the orders of magnitude let us consider the model where $\gamma_{\vec{q}}^{(3)} = \gamma_{\vec{q}}^{(4)} = \omega_{\vec{q}}$ in Eq. (6). Then Eqs. (8) and (33) with (16) and (17) imply $a(T\to\infty) = A_0(\vec{q}, T\to\infty) = 2k_BT$, so that Eqs. (40)–(43) lead to $2k_B\theta_1 = a(0) = c_1/\lambda_4$ and $2k_B\theta_2 = 1/\lambda_4 = g/c_2$. Since $g = 4(\lambda_3/\lambda_4)^2$, we find with (46) that $T_c/\theta_2 = c_2 - 1 = 4(\lambda_3^2/\lambda_4) - 1$. The assumption $T_c/\theta_2 \gg 1$ together with (49) then imply that $\epsilon^* \ll 1$, which is compatible, e.g., with the temperature $T^* = 120\,\mathrm{K}$ of Fig. 1.

We next derive an expression for $\omega_0(\vec{q}, T)$ away from $\vec{q}_R$. Since $2\vec{q}_R$ is a reciprocal lattice vector we have $\omega_{\vec{q}_R - \vec{q}} = \omega_{\vec{q}_R + \vec{q}}$ and $\gamma^{(n)}_{\vec{q}_R - \vec{q}} = \gamma^{(n)}_{\vec{q}_R + \vec{q}}$ so that because of (1), (16), and (17) ρ has the same property, and, according to (23), A_n does too. Therefore, Eq. (26) also implies

$$\omega_0^2(\vec{q}_R - \vec{q}, T) = \omega_0^2(\vec{q}_R + \vec{q}, T) \quad (52)$$

and a Taylor-series expansion in $\vec{q}$, combined with (50), leads to the general form

$$\omega_0^2(\vec{q}_R - \vec{q}, T) = \alpha_0\epsilon + \beta_0(\hat{q})\, q^2 + O(q^4, \epsilon q^2) , \quad (53)$$

where $\hat{q} = \vec{q}/q$, $\alpha_0 = \omega_{\vec{q}_R}^2/\epsilon^*$. This shows that at $T = T^*$ the renormalization is negligible, $\omega_0 \cong \omega_{\vec{q}}$, at least not too far away from $\vec{q}_R$. In particular, Eq. (53) gives a horizontal tangent of ω_0 at $\vec{q}_R$ which, for $T = T^*$, is in agreement with Fig. 1.

V. CENTRAL PEAK AND EPR LINEWIDTH

From Eq. (25) the height of the central peak is found to be

$$S(\vec{q}, 0) \propto \Gamma(0)\omega_0^{-4} , \quad (54)$$

which according to (53) diverges as ϵ^{-2} at $\vec{q} = \vec{q}_R$. Its width, defined by $S(\vec{q}, \Delta\omega) = \frac{1}{2}S(\vec{q}, 0)$,[10] is

$$\Delta\omega = \omega_0^2/\Gamma(0) \quad (55)$$

and goes to zero as ϵ at $\vec{q} = \vec{q}_R$. For this result it is essential that

$$\Gamma(0) = \delta^2/\gamma = \lambda_4^2 g \omega_{\vec{q}} \gamma_{\vec{q}}^{(4)} A_1(1 + \lambda_4 A_0)^{-2} \quad (56)$$

is finite for $\vec{q} \cong \vec{q}_R$ and $T \gtrsim T_c$. This depends crucially on A_1 or, provided that the limit $\Gamma_0 \to 0$ of Eq. (33) is legitimate, on $\rho(\vec{q}, 0)$. As we saw in Sec. II, $\rho_1 = 0$ for $\omega < \omega_{\vec{q}_R}$ and $\rho_0 = 0$ for $\omega < \Omega_{\vec{q}}^-$. Now, in the case when the two branches in Fig. 1 do not touch, $\Omega_{\vec{q}_R}^- > 0$ and Eq. (33) gives us $A_1(\vec{q}_R, T) = 0$, while for $\Gamma_0 \neq 0$ we may still get $A_1(\vec{q}_R, T) \neq 0$, according to (23). Thus, ρ_1 is of no consequence for the existence of a central peak near $\vec{q}_R$, but ρ_0 is crucial and the central peak depends sensitively on the relative magnitude of $\Omega_{\vec{q}_R}^-$ and Γ_0. So we have to evaluate A_1 more carefully.

Near $\omega = \Omega_{\vec{q}}^-$ the main contributions to the $\vec{k}$ sum in (1) come from the neighborhood of the minimum of $\omega_{\vec{q}-\vec{k}} - \omega_{\vec{k}}$. The latter has the form

$$\omega_{\vec{q}-\vec{k}} - \omega_{\vec{k}} = \Omega_{\vec{q}}^- + b_{\vec{q}}(\vec{k} - \vec{k}_{\vec{q}})^2 , \quad (57)$$

where the position $\vec{k}_{\vec{q}}$ of the minimum is fairly close to the center of the Brillouin zone (see Fig. 1) and $b_{\vec{q}}$ will in general depend on the direction of $\vec{k} - \vec{k}_{\vec{q}}$. Inserting (57) and (16) into (1) yields, for $\omega \cong \Omega_{\vec{q}}^-$, $|\Omega_{\vec{q}}^-| \ll \omega_{\vec{k}\vec{q}} \ll k_BT$,

$$\rho_0(\vec{q}, \omega) = B_{\vec{q}}(2\pi)^{-1} \int d^3k'\, \delta(\omega - \Omega_{\vec{q}}^- - b_{\vec{q}}(\hat{k}')k'^2) , \quad (58)$$

where

$$B_{\vec{q}} = (2\pi)^{-2}(V/N)k_BT\gamma^{(4)}_{\vec{k}_{\vec{q}}}\gamma^{(4)}_{\vec{q}-\vec{k}_{\vec{q}}}\,\omega^{-2}_{\vec{k}_{\vec{q}}} . \quad (59)$$

Evaluating (58) we find

$$\rho_0(\vec{q}, \omega) = B_{\vec{q}}\langle b_{\vec{q}}^{-3/2}\rangle (\omega - \Omega_{\vec{q}}^-)^{1/2}\theta(\omega - \Omega_{\vec{q}}^-) , \quad (60)$$

where the brackets $\langle\ \rangle$ mean average over the direction $\hat{k}'$.

Inserting (60) into (23) yields

$$A_1 = \pi B_{\vec{q}}\langle b_{\vec{q}}^{-3/2}\rangle (\tfrac{1}{2}\Gamma_0)^{1/2} I(\Omega_{\vec{q}}^-/\Gamma_0) , \quad (61)$$

where

$$I(\nu) \equiv \frac{2^{5/2}}{\pi}\int_0^\infty dx \left(\frac{x(x^2+\nu)}{1+(x^2+\nu)^2}\right)^2 = [-\nu + (\nu^2+1)^{1/2}]^{1/2} + \tfrac{1}{2}(\nu^2+1)^{-1/2}[-\nu + (\nu^2+1)^{1/2}]^{-1/2} . \quad (62)$$

$I(\nu)$ is a positive monotonically decreasing function with the asymptotic behavior

$$I(\nu) = \begin{cases} (2|\nu|)^{1/2} , & \nu \to -\infty \\ (2/\nu)^{1/2} , & \nu \to +\infty . \end{cases} \quad (63)$$

From (60), (61), and (63) we immediately recover the general result (33) for the case $\Gamma_0 \to 0$, which vanishes for $\Omega_{\vec{q}}^- \geq 0$. This shows that in order to have a central peak at $\vec{q}_R$, in the case where there is no crossing of the two branches of Fig. 1, Γ_0 must be of the order of the minimal distance $\Omega_{\vec{q}_R}^-$ of these branches. This is quite satisfactory from the point of view of the self-consistency conditions (38), the second of which requiring, according to (39), that $\frac{1}{4}\Gamma_0^2$ be small while the first demands that Γ_0 is nonzero.

A more quantitative evaluation of Eq. (56) confirms this observation. In the model $\gamma_{\vec{q}}^{(4)}=\omega_{\vec{q}}$ used earlier we have for $T=T^*$ that $\lambda_4^2 g\omega_{\vec{q}}\gamma_{\vec{q}}^{(4)}(1+\lambda_4 A_0)^{-2}=\omega_{\vec{q}}^2/(2k_B T^*)$. Since $V/N=a^3=(\pi\sqrt{3}/q_R)^3$, expression (59) is $B_{\vec{q}}=\pi 3^{3/2}k_B T^*/(4q_R^3)$. Assuming $|\Omega_{\vec{q}_R}|/\Gamma_0 \lesssim 1$, so that according to (62) $I\cong I(0)=\frac{3}{2}$, inserting (61) and the above estimates into (56) yields, for $\vec{q}\cong\vec{q}_R$,

$$\Gamma(0)\cong\tfrac{9}{16}\sqrt{\tfrac{3}{2}}\,\pi^2\langle b_{\vec{q}}^{-3/2}\rangle\,\omega_{\vec{q}_R}^2\,q_R^{-3}\Gamma_0^{1/2}\ . \tag{64}$$

$b_{\vec{q}_R}$ can be determined from (57) and from the fact that for small k one has $\omega_{\vec{k}}=ck$. For $k\to 0$ and $\vec{q}=\vec{q}_R$, (57) is still a reasonable approximation and reads

$$\omega_{\vec{q}_R}-\Omega_{\vec{q}_R}\cong b_{\vec{q}_R}k_{\vec{q}_R}^2\ . \tag{65}$$

The derivative with respect to $\vec{k}$ of (57) yields for these values of $\vec{k}$ and $\vec{q}$

$$\left(\frac{\partial\omega_{\vec{q}_R-\vec{k}}}{\partial\vec{k}}\right)_{k=0}-c\hat{k}\cong-2b_{\vec{q}_R}\vec{k}_{\vec{q}_R}\ , \tag{66}$$

but according to (53) (see also Fig. 1), $(\partial\omega_{\vec{q}_R-\vec{k}}/\partial\vec{k})_{k=0}=0$. Since $|\Omega_{\vec{q}_R}|\ll\omega_{\vec{q}_R}$ (see Fig. 1), we have from (65) and (66) $k_{\vec{q}_R}\cong 2\omega_{\vec{q}_R}/c$ and

$$b_{\vec{q}_R}\cong c^2/4\omega_{\vec{q}_R}\ . \tag{67}$$

Inserting this into (64) yields, combined with the first condition (38) and neglecting the anisotropy of the phonon velocity c,

$$(\Gamma_0/\omega_{\vec{q}_R})^{1/2}\cong\tfrac{9}{2}\sqrt{\tfrac{3}{2}}\,\pi^2(\omega_{\vec{q}_R}/cq_R)^3\ . \tag{68}$$

According to Fig. 1, $\omega_{\vec{q}_R}\ll cq_R$, which leads us to conclude that $\Gamma_0\cong\Gamma(0)\ll\omega_{\vec{q}_R}$, and hence that the second condition (38) is well satisfied. But in addition we see from (55) that at $T=T^*>T_c$ the central peak is completely washed out over the entire soft-mode peak, $\Delta\omega/\omega_0=\omega_0/\Gamma_0\gg 1$, in agreement with observation.[4]

Finally, we calculate the EPR linewidth ΔH which is obtained from[9,11]

$$\Delta H\propto(1/N)\sum_{\vec{q}}S(\vec{q},\,0)\ . \tag{69}$$

Making use of (53) and describing the anisotropy around $\vec{q}_R$ in the form[9]

$$\beta_0(\hat{q})=[1-(1-\Delta)\zeta^2]\beta_0\ , \tag{70}$$

where $\zeta=\hat{q}\cdot\hat{q}_R$, the leading singularity of ΔH is contained in

$$(\Delta H)_{\text{sing}}\propto\int_0^1 d\zeta\int_0^{q_m(\zeta)}q^2\,dq\times\{\alpha_0\epsilon+\beta_0[1-(1-\Delta)\zeta^2]\,q^2\}^{-2}\ , \tag{71}$$

where $q_m(\zeta)$ is an angle-dependent cutoff. In the case that $q_m(\zeta)\neq 0$, for all angles, one finds

$$(\Delta H)_{\text{sing}}\propto\epsilon^{-\nu}\ , \tag{72}$$

with

$$\nu=\tfrac{1}{2}\quad\text{if }0<\Delta\lesssim 1\ . \tag{73}$$

In the "two-dimensional" case[10] $\Delta=0$, one has instead of (71)

$$(\Delta H)_{\text{sing}}\propto\int_0^{q_{\parallel m}}dq_\parallel\int_0^{q_{\perp m}}q_\perp\,dq_\perp[\alpha_0\epsilon+\beta_0 q_\perp^2]^{-2}\ , \tag{74}$$

where $q_\perp=(1-\zeta^2)^{1/2}q$. Here the result is

$$\nu=1\quad\text{if }\Delta=0\ . \tag{75}$$

These values of the exponent ν are to be expected in an ϵ domain where critical fluctuations are unimportant. In comparing this result with the exponent found in Ref. 11, one should bear in mind that this comparison only holds for the "fast-fluctuation" regime valid for $\epsilon\gtrsim 0.01$. From Fig. 2 of Ref. 11 one deduces $\nu\cong\frac{2}{3}$ for $\epsilon\cong 0.01$ ($T\cong 106.7$ K) and $\nu\cong 1.0$ for $\epsilon\cong 0.09$ ($T\cong 115$ K). [Ncte that ν is obtained from the slope in Fig. 2 of Ref. 11 by addition of $\epsilon(1+\epsilon)^{-1}$.] We conclude that for $107\lesssim T\lesssim 110$ K the exponent ν is determined by the ("fast") critical fluctuations ($\nu\cong\frac{2}{3}$), while for $T\cong 115$ K the critical fluctuations are negligible and ν is determined by the mean-field value 1.0 valid for the "two-dimensional" case[10] $\Delta=0$. This is in fair agreement with the experimental value $\Delta=0.017\pm 0.010$ obtained in Ref. 11.

VI. CONCLUSION

Critical fluctuations have been neglected in the present work. In a lattice-dynamical treatment they are essentially described by the ladder insertion of the diagram considered by the authors of Ref. 8. This diagram has the form of mode-mode coupling theory and therefore is expected to be important in the critical region. However, no critical exponents have as yet been obtained in this way. Our purpose was to show that in the noncritical ϵ domain above[15] T_c the important diagrams are the chains which lead to a consistent description of all the points enumerated in Sec. I but with a real harmonic frequency.

The most interesting feature resulting from our theory is the occurrence of a central peak $S(\vec{q},\,0)$. With Eqs. (54), (56), (22), and (11) this central peak can be traced back to the propagator G of the collective coordinate $\epsilon_{\vec{q}}$ of Eq. (13). Since $\epsilon_{\vec{q}}$ is essentially the energy density, we conclude that the central peak can be understood as occurring from energy fluctuations. This gives support to Feder's theory,[7,10] with the important qualification, however, that for $T>T_c$ it is the extreme short-wave part $\vec{q}\cong\vec{q}_R$ of $\epsilon_{\vec{q}}$ that is important. This means that a thermodynamic treatment[7] leading to a Landau–Placzek peak is not applicable. However Feder's theory[7] can be understood as an "analytic" continuation from a hydrodynamic $\vec{q}\cong 0$ to $\vec{q}\cong\vec{q}_R$. It is also in this sense that the frequency ω_A of Schneider[10] is justly called "adiabatic." Indeed the adiabatic limit is defined as "first $q\to 0$ then ω

$\to 0$"[20] such that, in Schneider's notation,[10] $\omega_A(\vec{q}) \gg \lambda_2(\vec{q})$, λ_2 being the damping. If now $\vec{q}$ is continued from $\vec{q} \cong 0$ to $\vec{q} \cong \vec{q}_R$ this inequality is preserved so that ω_A becomes high frequency.

The important fact giving rise to a central peak in our theory was recognized to be the quasitouching behavior of the two halves of the lowest transverse-phonon branch exhibited in Fig. 1. This led us to conclude that the two-phonon density of states $\rho_0(\vec{q}, \omega)$ of Eq. (1) is nonzero for the relevant values $\vec{q} \cong \vec{q}_R$, $\omega \cong 0$ and, consequently, gives rise to a dominance of the chain diagrams. Of course, other models[21] may also give rise to a central peak.

Indeed, Cowley[21] has obtained a central peak at $\vec{q} = 0$ from the simple bubble diagram with the same phonon of finite width occurring in both lines. As Cowley points out, this diagram would vanish at $\vec{q} = 0$ for a nonpiezoelectric material, such as $SrTiO_4$, owing to the symmetry of the three-phonon vertex; for $\vec{q} \cong \vec{q}_R$ this is, of course, not the case. It is interesting that Cowley obtains the same temperature dependence $\sim \epsilon^{-2}$ of the central peak as in our Eq. (54).

In Silberglitt's treatment,[21] one line of the two-phonon bubble is replaced by a pair of lines which is treated self-consistently and therefore, presumably, reproduces part of the ladder insertions of Ref. 8. This "dressed" phonon pair therefore also represents a collective mode which is of the type of second sound[8] because it has long wavelength. In this terminology our short-wavelength collective mode $\epsilon_{\vec{q}}$ may be called "collisionless," i.e., nonhydrodynamic second sound, an interpretation which has been proposed earlier.[17,18]

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APPENDIX: THE PYTTE–FEDER INTERACTION

Pytte and Feder's interaction Hamiltonian, Eq. (16) of Ref. 1, can be written in the form

$$H_4 = (1/N) \sum_{\vec{q}_1 \cdots \vec{q}_4} \sum_{\lambda\lambda'} \Gamma^{(4)}_{\lambda\lambda'}(\vec{q}_1 \cdots \vec{q}_4) \times R_\lambda(\vec{q}_1) R_\lambda(\vec{q}_2) R_{\lambda'}(\vec{q}_3) R_{\lambda'}(\vec{q}_4) \Delta\left(\sum_{S=1}^{4} \vec{q}_S\right), \quad (A1)$$

where

$$\vec{R}(\vec{q}) = \langle \vec{R}(\vec{q}) \rangle + \sum_\lambda \vec{e}(\lambda, \vec{q}) \omega_{\lambda\vec{q}}^{-1/2} Q_{\lambda\vec{q}} \quad (A2)$$

is the Fourier transform of $\vec{R}(\vec{l})$ of Ref. 1. The normal coordinates $Q_{\lambda\vec{q}}$ and polarization vectors $\vec{e}(\lambda, \vec{q})$ are determined by the diagonalization of the harmonic Hamiltonian $H_0 = T + H_2$ [Eqs. (4) and (11) of Ref. 1],

$$H_0 = \frac{1}{2} \sum_{\lambda\vec{q}} \omega_{\lambda\vec{q}} \{ P_{\lambda\vec{q}} P^\dagger_{\lambda\vec{q}} + Q_{\lambda\vec{q}} Q^\dagger_{\lambda\vec{q}} \} + \text{terms in } \langle \vec{R}(\vec{q}) \rangle, \quad (A3)$$

where $P_{\lambda\vec{q}}$ is the canonically conjugate of $Q_{\lambda\vec{q}}$ defined by

$$i[H_0, Q_{\lambda\vec{q}}] = \omega_{\lambda\vec{q}} P^\dagger_{\lambda\vec{q}}. \quad (A4)$$

The unitary diagonalizing matrix $e_\lambda(\lambda', \vec{q})$ is determined by the equations

$$e_\lambda(\mu, \vec{q}) \theta_\lambda(\vec{q}) = e_\lambda(\mu, \vec{q}), \qquad \lambda, \mu = 1, 2, 3 \quad (A5)$$

$$\sum_{\lambda'} [v_{\lambda'\lambda}(\vec{q}) - \omega^2_{\mu\vec{q}} \delta_{\lambda'\lambda}] e_{\lambda'}(\mu, \vec{q}) = 0, \quad (A6)$$

where

$$\theta_{\lambda\lambda'}(\vec{q}) = \theta_\lambda(\vec{q}) \delta_{\lambda\lambda'} \quad (A7)$$

and $v_{\lambda\lambda'}(\vec{q})$ are the Fourier transforms of the matrices in Eqs. (4) and (11) of Ref. 1, respectively, and (A7) is explicitly given by Eq. (10) of Ref. 1. We note that Eq. (A5) implies $\theta_\lambda(\vec{q}) = 1$ for all λ, $\vec{q}$, so that $T = \frac{1}{2}\sum_{\lambda\vec{l}} R^2_\lambda(\vec{l})$ is local in $\vec{l}$, whereas in the coordinates of Ref. 1 T is nonlocal in $\vec{l}$.

The vertex function in Eq. (A1) is defined by

$$4\Gamma^{(4)}_{\lambda\lambda'}(\vec{q}_1 \cdots \vec{q}_4) = 2\Gamma_{\lambda\lambda'}(0) - 2\sum_{S=1}^{4} \Gamma_{\lambda\lambda'}(\vec{q}_S) + \sum_{S<S'} \Gamma_{\lambda\lambda'}(\vec{q}_S + \vec{q}_{S'}), \quad (A8)$$

where $\Gamma_{\lambda\lambda'}(\vec{q})$ are the elements of the matrix defined in Eq. (19) of Ref. 1. Making use of the trigonometric identity

$$2\cos 0 - 2\sum_{S=1}^{4} \cos\alpha_S + \sum_{S<S'} \cos(\alpha_S + \alpha_{S'}) = 8 \prod_{S=1}^{4} e^{i\alpha_S/2} \sin\tfrac{1}{2}\alpha_S, \quad (A9)$$

where $\sum_{S=1}^{4} \alpha_S = \pm 2\pi n$, n being an integer, the particular form of the matrix $\Gamma(\vec{q})$ implies that Eq. (A8) takes the form

$$8\Gamma^{(4)}_{11}(\vec{q}_1 \cdots \vec{q}_4) = \Gamma_1 \left(\prod_{S=1}^{4} \gamma_2(\vec{q}_S) + \prod_{S=1}^{4} \gamma_3(\vec{q}_S) \right),$$

$$8\Gamma^{(4)}_{12}(\vec{q}_1 \cdots \vec{q}_4) = 2\Gamma_2 \prod_{S=1}^{4} \gamma_3(\vec{q}_S), \quad (A10)$$

etc., where Γ_1 and Γ_2 are defined in Eq. (18) of Ref. 1 and

$$\gamma_\lambda(\vec{q}) = e^{i\vec{q}\cdot\vec{\xi}_\lambda/2} \sin\tfrac{1}{2}\vec{q}\cdot\vec{\xi}_\lambda, \quad (A11)$$

with[1] $\vec{\xi}_1 = a(1, 0, 0)$, etc.

In the particular case where the unrenormalized frequency $\omega_{\lambda\vec{q}}$ is independent of λ, we have from (A6)

$$e_\lambda(\lambda', \vec{q}) = \delta_{\lambda\lambda'} , \quad v_{\lambda\lambda'}(\vec{q}) = \omega^2_{\vec{q}}\delta_{\lambda\lambda'} . \tag{A12}$$

If we further put $\Gamma_2 = 0$ in (A10) and assume $T > T_c$ so that $\langle \vec{R}(\vec{q}) \rangle = 0$ in (A2), Eq. (A1) becomes a sum of three terms:

$$H_{4\lambda} = (\tfrac{1}{8}\Gamma_1) \frac{1}{N} \sum_{\vec{q}_1 \cdots \vec{q}_4} \left(\sum_{\mu \neq \lambda} \prod_{S=1}^{4} \omega^{-1/2}_{\vec{q}_S} \gamma_\mu(\vec{q}_\cdot) \right) \times Q_{\lambda\vec{q}_1} \cdots Q_{\lambda\vec{q}_4} \Delta\left(\sum_{S=1}^{4} \vec{q}_S \right) . \tag{A13}$$

On restricting the $\vec{q}$'s to the body-diagonals $\vec{q} = q(\pm 1, \pm 1, \pm 1)$, $q \leq \pi/a$, $H_{4\lambda}$ reduces to our Eq. (6) for H_4 with $\lambda_4 = 12\Gamma_1$, $\omega_{\vec{q}}\gamma^{(4)}_{\vec{q}} = e^{\pm iqa} \sin^2 \frac{1}{2}qa$ and $Q_{\lambda\vec{q}} \to Q_{\vec{q}}$ (the factors $e^{\pm iqa}$ only contribute a sign for umklapp processes).

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