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Collective modes in charge-density waves and long-range Coulomb interactions

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We study theoretically the collective modes in charge-density waves in the presence of long-range Coulomb interaction. We find that earlier works by Takada and his collaborators are inadequate since they introduced inconsistent approximations in evaluating a variety of correlation functions. The amplitude mode is unaffected by the Coulomb interaction, while the phase mode splits into the phason with linear dispersion (i.e., acoustic mode) and the optical mode with an energy gap in the presence of the Coulomb interaction. In particular, we establish the temperature dependence of the phason velocity v_ϕ. A comparison with recent neutron-scattering data on the phason velocity in the charge-density wave of a single crystal of blue bronze $K_{0.3}MoO_3$ indicates that mean-field theory which includes the long-range Coulomb interaction gives an excellent description of the observed phason velocity.

I. INTRODUCTION

Fröhlich conduction in the charge-density wave (CDW) in $NbSe_3$ and other systems is now well established both experimentally and theoretically.[1] More recently the validity of three-dimensional 3D mean-field theory has been established in most of these systems. For example, the ratio of Δ_a/T_c observed in most CDW's is, we believe, due to imperfect nesting.[2] Here Δ_a is the quasiparticle energy gap determined from the electric resistance and T_c is the CDW transition temperature. Perhaps the clearest signature of imperfect nesting is the pressure dependence of T_c. In the model with imperfect nesting, T_c decreases with pressure since the pressure increases the dimensionality.[2] On the other hand, in an alternative fluctuation model, T_c should increase with pressure, since the fluctuation is suppressed by emerging dimensionality.[3,4] However, the fluctuation effects on the thermodynamics and the transport properties are understood only partially.[5]

The object of this paper is to study the role of long-range Coulomb interaction on collective modes of the CDW.[6] This problem has been considered by Lee and Fukuyama[7] at $T=0$ K and later studied more extensively by Takada and his collaborators[8] for all temperatures. However, unfortunately due to their inconsistent approximations [i.e., some correlation functions are evaluated in the static limit ($\omega \ll \zeta$), while others are in the dynamic limit ($\omega \gg \zeta$) where ω is the frequency and $\zeta = v_F q$ with q the wave vector in the most conducting direction], their results are not reliable especially in the vicinity of $T=T_c$. Within the Fröhlich model[9] all the functions necessary for the present study are already available[10] for arbitrary α ($=\omega/\zeta$), when the effect of imperfect nesting is neglected. As we shall see, the condensate density for arbitrary α plays an important role in the present analysis. Then within the time-dependent mean-field theory (i.e., the random-phase approximation) we determine the collective modes' dispersion at all temperatures. The spectral weights of these modes, which are proportional to the scattering amplitude of neutrons, are obtained. At high temperatures the acoustic mode almost exhausts the phason spectral weight, while at low temperatures (say $T \le 0,2T_c$) the optical mode does. The present theory describes extremely well the temperature dependence of the phason velocity v_ϕ observed[11] in a single crystal of $K_{0.3}MoO_3$ by neutron scattering. This indicates again that the effect of fluctuations is rather small even in CDW of blue bronze.

II. FLUCTUATION PROPAGATORS

We consider the Fröhlich Hamiltonian[9] properly generalized to three dimensions and we add the long-range Coulomb interaction

$$H_c = 4\pi e^2 \sum_q \frac{1}{q^2} n_q n_{-q} , \tag{1}$$

where n_q is the electron-density operator. Then the amplitudon and phason dispersions are determined[9] from the pole of

$$\begin{aligned} D_A(q,\omega) &= \{1 - g^2\omega_Q^2(\omega_Q^2-\omega^2)^{-1}\langle[\Delta_1,\Delta_1]\rangle\}^{-1} \\ &\quad \times \omega_Q^2(\omega_Q^2-\omega^2)^{-1} \\ &= -(2\Delta)^2(\lambda f)^{-1}\left[\frac{m^*}{m}\omega^2 - \zeta^2 - (2\Delta)^2\right]^{-1} \end{aligned} \tag{2}$$

and

$$D_\phi(q,\omega)=\{1-g^2\omega_Q^2(\omega_Q^2-\omega^2)^{-1}\langle[\Delta_2,\Delta_2]\rangle\}^{-1}\omega_Q^2(\omega_Q^2-\omega^2)^{-1}$$
$$=-(2\Delta)^2(\lambda f)^{-1}\left[\frac{m^*}{m}\omega^2-\zeta^2-\omega_p^2 f\left[1-\frac{\omega_p^2(1-f)}{\omega^2-\zeta^2}\right]^{-1}\right]^{-1}$$
$$\cong-(2\Delta)^2(\lambda f)^{-1}(1-f)\left[\left[\frac{m^*}{m}(1-f)+f\right]\omega^2-\zeta^2\right]^{-1}, \tag{3}$$

where Δ_1 and Δ_2 are the real and the imaginary parts of the CDW order parameter and the effect of the Coulomb interaction is incorporated within the mean-field theory.[12] The last line in Eq. (3) is obtained by putting $\omega_p=\infty$. Further, here we limit ourselves to the longitudinal case $\mathbf{q}\|\mathbf{a}$ the most conducting direction. A short derivation of Eq. (3) is sketched in Appendix A. Here λ is the dimensionless electron-phonon coupling constant, $\omega_p=(4\pi e^2 n/m)^{1/2}$ the plasma frequency, and f is the generalized condensate density[10]

$$f=\begin{cases}-\frac{1}{2}\int_{-\infty}^{\infty}d\phi\tanh(\frac{1}{2}\beta\Delta\cosh\phi)[\sinh^2(\phi-\phi_0)-(1-\alpha^2)\tilde{\zeta}^2]^{-1} & \text{for } \alpha\le 1 \quad (4)\\ \frac{1}{2}\int_{-\infty}^{\infty}d\phi\tanh(\frac{1}{2}\beta\Delta\cosh\phi)[\cosh^2(\phi-\phi_0)+(1-\alpha^2)\tilde{\zeta}^2]^{-1} & \text{for } \alpha\ge 1 \quad (5)\end{cases}$$

and

$$\tanh\phi_0=\begin{cases}\alpha & \text{for } \alpha<1\\ \alpha^{-1} & \text{for } \alpha>1\,,\end{cases} \tag{6}$$

respectively, and $\tilde{\zeta}=\zeta/[2\Delta(T)]$. Here we neglect the effect of imperfect nesting for simplicity. Further,

$$f=1 \quad \text{for } \alpha=1 \tag{7}$$

independent of temperature.

In the adiabatic limit [i.e., $\omega,\zeta\ll 2\Delta(T)$] we obtain

$$f_s=\lim_{\zeta\to 0}\lim_{\omega\to 0}=2\pi T\sum_{n=0}^{\infty}\frac{\Delta^2}{(\omega_n^2+\Delta^2)^{3/2}} \tag{8}$$

and

$$f_d=\lim_{\omega\to 0}\lim_{\zeta\to 0}\int_0^{\infty}d\phi\,\mathrm{sech}^2\phi\tanh(\tfrac{1}{2}\beta\Delta\cosh\phi)\,. \tag{9}$$

Here subscripts s and d mean the static and the dynamic limit.

The temperature dependence of f_s and f_d is shown in Fig. 1. From Eq. (2) the dispersion of the amplitudon is given by

$$\omega_A^2=\frac{m}{m^*}[(2\Delta)^2+\zeta^2]\cong\lambda\omega_Q^2 f\left|+\frac{\zeta^2}{(2\Delta)^2}\right|, \tag{10}$$

where the second line is valid as long as $\lambda^{1/2}\omega_Q<2\Delta(T)$, which holds always except in the immediate vicinity of $T=T_c$ (i.e., $T_c-T\le 10^{-2}T_c$). Here the phason mass m^* is given by

$$\frac{m^*}{m}=1+(2\Delta)^2/\lambda\omega_Q^2 f\,. \tag{11}$$

It is of interest to note that Eq. (10) together with Eq. (11) guarantees a solution $\omega^2=\zeta^2=\lambda\omega_Q^2$ in the CDW independent of temperature.

At $T=0$ K, we expand f in powers of $\zeta/2\Delta$ and $\omega/2\Delta$ as

$$f=1+\frac{2}{3}\frac{\omega^2-\zeta^2}{(2\Delta)^2}+\frac{8}{15}\frac{(\omega^2-\zeta^2)^2}{(2\Delta)^4}+\cdots \tag{12}$$

and we obtain[9,13]

$$\omega_A^2=\lambda\omega_Q^2+\frac{1}{3}\frac{m}{m^*}\zeta^2\,. \tag{13}$$

For $T>0$ K, it is important to distinguish two limits ($\omega\gg\zeta$ and $\omega\ll\zeta$), since the latter limit is most likely realized in the neutron-scattering experiment.[14,15] Especially in the high-temperature regime we obtain

$$\omega_A^2=\begin{cases}\lambda\omega_Q^2 f_d+\frac{1}{2}\zeta^2 & \text{for } \omega\gg\zeta\\ \lambda\omega_Q^2 f_s+\dfrac{m}{m_s^*}\zeta^2 & \text{for } \omega\ll\zeta\,.\end{cases} \tag{14}$$

Therefore the amplitude frequency $\omega_A=\sqrt{\lambda}\omega_Q(f_s)^{1/2}$ obtained by Rice and Strassler[15] is valid only in the static limit. Here we introduced $m_s^*(T)$, which is defined by

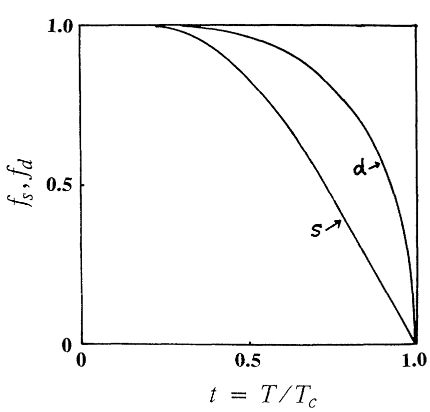

FIG. 1. The condensate density f_s (the static limit) and f_d (the dynamic limit) are evaluated numerically and shown as a function of the reduced temperature $t=T/T_c$.

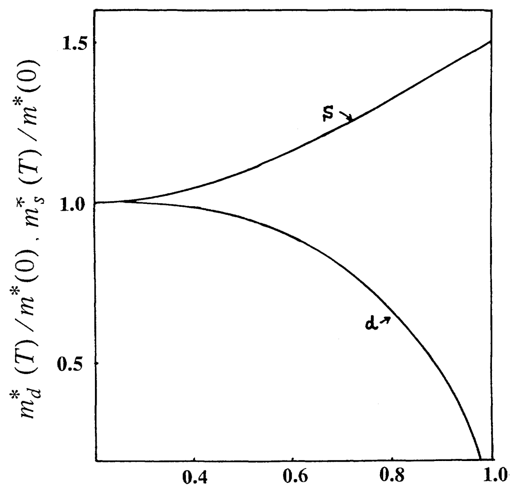

FIG. 2. The phason mass $m_s^*(T)$ (in the static limit) and $m_d^*(T)$ (the dynamic limit) are shown as a function of the reduced temperature.

$$\frac{m_s^*(T)}{m} = 1 + \frac{(2\Delta)^2}{\lambda\omega_Q^2 f_s} ; \tag{15}$$

similarly we can define $m_d^*(T)$ by replacing f_s in Eq. (15) by f_d. The temperature dependences of $m_s^*(T)$ and $m_d^*(T)$ are shown in Fig. 2. It is important to note that the temperature dependence of $\omega_A(T)$ is quite different especially in the vicinity of $T = T_c$ depending on which limit you are in.

III. ACOUSTIC AND OPTICAL MODES

As already pointed out by Takada and co-workers,[8] the longitudinal phason consists of two modes, which are determined from

$$\left[1 + \frac{(2\Delta)^2}{\lambda\omega_Q^2}(f^{-1} - 1)\right]\omega^2 = \xi^2 . \tag{16}$$

We shall examine these modes here.

A. Acoustic mode

One solution is given by

$$\omega^2 = \frac{m}{m^*}\left[1 - f + \frac{m}{m^*} f\right]^{-1} \xi^2 , \tag{17}$$

where m^*/m defined in Eq. (11) has to be used. For not too low temperatures ($T \geq 0.3T_c$) we have $\omega \ll \xi$ and we can simplify Eq. (17) as

$$\omega = v_\phi q , \tag{18}$$

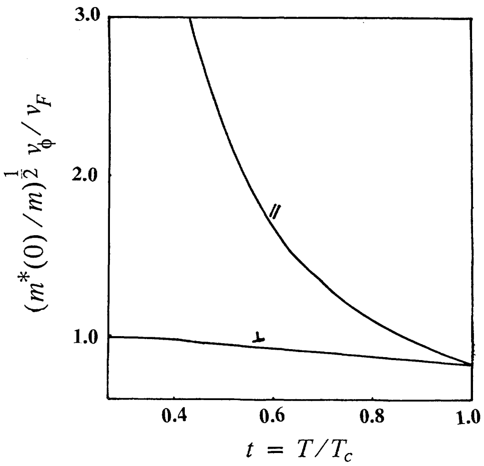

FIG. 3. The longitudinal and the transverse phason velocity v_ϕ and $v_{\phi\perp}$ are shown as a function of the reduced temperature.

with

$$v_\phi / v_F = \left[\frac{m}{m_s^*}\right]^{1/2} \left[1 - f_s + \frac{m}{m_s^*} f_s\right]^{-1/2} . \tag{19}$$

The temperature dependence of v_ϕ / v_F is shown in Fig. 3. As T decreases from T_c, v_ϕ increases rapidly and ultimately at low temperatures ($T \cong 0.3T_c$) the static limit becomes no longer valid. Also as discussed already by Takada and this collaborators[8] the acoustic mode merges with another acoustic mode around ($T \cong 0.2T_c$) and completely disappears for the lower temperatures. It is of interest to consider the spectral weight of the acoustic model. From Eq. (3) we obtain

$$\int_0^\infty d\omega\, \omega\, \mathrm{Im} D_\phi(q,\omega) = \frac{\pi}{2}\omega_Q^2 , \tag{20}$$

which in independent of ξ, T, and ω_p. Now if we evaluate the contribution of the acoustic pole we obtain

$$\int_0^\infty d\omega\, \omega\, \mathrm{Im} D_\phi^{\mathrm{ac}}(q,\omega) = \frac{\pi}{2}\omega_Q^2 \left[\frac{1 - f_s}{1 - f_s + \frac{m}{m_s^*} f_s}\right] \times \left[1 - \frac{m}{m_s^*}\right] . \tag{21}$$

The acoustic mode almost exhausts the spectral weight as long as $1 - f_s \gg m/m_s^*$ (i.e., $T \geq 0.3T_c$).

It is also possible to derive the acoustic mode for an arbitrary $\mathbf{q}$. Making use of the angular integral

$$\frac{1}{2\pi}\int_0^{2\pi} d\chi \frac{(\xi_\parallel + \sqrt{2}\xi_\perp \cos\chi)^2}{(\xi_\parallel + \sqrt{2}\xi_\perp \cos\chi)^2 - \omega^2} = 1 + \frac{\omega}{2}\{[(\xi_\parallel - \omega)^2 - 2\xi_\perp^2]^{-1/2} - [(\xi_\parallel + \omega)^2 - 2\xi_\perp^2]^{1/2}\}$$
$$\cong 1 + \omega^2 \xi_\parallel (\xi_\parallel^2 - 2\xi_\perp^2)^{-3/2} + \omega^4 \xi_\parallel (\xi_\parallel^2 - 3\xi_\perp^2)(\xi_\parallel^2 - 2\xi_\perp^2)^{-7/2} + \cdots \tag{22}$$

we obtain

$$\omega=\left[\frac{m}{m_s^*}\right]^{1/2}\left[\left[1-f_s+\frac{m}{m_s^*}f_s\zeta_\parallel^3(\zeta_\parallel^2-2\zeta_\perp^2)^{-3/2}\right]^{-1/2}[\zeta_\parallel^2+(1-f_s)\zeta_\perp^2]^{1/2}\right]$$
$$\cong\left[\frac{m}{m_s^*}\right]^{1/2}(1-f_s)^{-1/2}[\zeta_\parallel^2+(1-f_s)\zeta_\perp^2]^{1/2}\ , \tag{23}$$

where $\zeta_\parallel=v_F q_\parallel$, $\zeta_\perp=v_{F\perp}q_\perp$. Unfortunately the first equation in Eq. (23) breaks down around $\zeta_\parallel\equiv\sqrt{2}\zeta_\perp$. But Eq. (23) is correct in the limits $\zeta_\perp=0$ and $\zeta_\parallel=0$. Also the second line in Eq. (23) is adequate for practical purposes. In particular, when $q_\parallel=0$ we obtain

$$\omega=\left[\frac{m}{m_s^*}\right]^{1/2}\zeta_\perp\ . \tag{24}$$

The effect of the long-range Coulomb interaction disappears completely in the transverse limit. In Fig. 3 we show also $v_{\phi\perp}$ as a function of the reduced temperature $t=T/T_c$. $v_{\phi\perp}$ increases with decreasing temperatures, but very slowly.

B. Optical mode

Another solution of Eq. (16) is readily found at $T=0$ K as

$$\omega_{\text{op}}^2=\tfrac{3}{2}\lambda\omega_Q^2+\frac{1}{5}\frac{m}{m^*}\zeta^2\ , \tag{25}$$

where we inserted Eq. (12) in place of f. Although the optical frequency is known[8] already the coefficient of the ζ^2 term is new. At low temperatures the optical frequency is given by

$$\omega_{\text{op}}^2\cong\tfrac{3}{2}[\lambda\omega_Q^2+(2\Delta)^2(1-f_a)](1-\tfrac{3}{2}G_d)^{-1}\ , \tag{26}$$

where

$$G_d=2\int_0^\infty d\phi\,\text{sech}^4\phi(1+e^{\beta\Delta\cosh\phi})^{-1}\ . \tag{27}$$

The optical frequency is fairly constant at low temperatures but starts increasing rapidly around $T\cong 0.3T_c$, then takes a maximum value of $\sim\Delta(T)$ around $T\cong 0.9T_c$, and then decreases rapidly to

$$\omega_{\text{op}}^2=(2\Delta)^2\left[1-\left[\frac{\pi\Delta}{4T}\right]^2\left[1-\frac{\lambda\omega_Q^2}{(2\Delta)^2}\right]^2\right] \tag{28}$$

and hits $2\Delta(T)$ at $2\Delta(T)=\sqrt{\lambda}\omega_Q$ from below. There will be no optical mode in the region $2\Delta(T)\le\sqrt{\lambda}\omega_Q$. Further, $\omega_{\text{op}}(T)>\omega_A(T)$ for all temperatures. At low temperatures the spectral weight of the optical mode is given by

$$\int_0^\infty d\omega\,\omega\,\text{Im}D_\phi^{\text{op}}(\omega,q)=\frac{\pi}{2}\omega_Q^2(1+x_0+\Xi)(x_0+\Xi)\times(1-f_d+x_0+2\Xi)^{-1}\ , \tag{29}$$

where $x_0=\lambda\omega_Q^2/(2\Delta)^2\cong m/m^*$,

$$\Xi=\tfrac{6}{5}(1-f_d+x_0)^2(1-\tfrac{3}{2}G_d)^{-2}\ . \tag{30}$$

The optical mode almost exhausts the spectral weight at low temperatures ($T\le 0.2T_c$). When $1-f_d\cong m/m^*$ the spectral weight is very quickly transferred to the acoustic mode as we have shown already. In Appendix B we derive the optical mode for an arbitrary **q** at $T=0$ K.

IV. CONCLUDING REMARKS

We have studied the collective mode in the CDW in the presence of the long-range Coulomb interaction. Although the amplitudon is unaffected by the Coulomb interaction we obtain results as to the dispersion of the amplitudon in the vicinity of $T=T_c$. The phason splits into two modes in the presence of the Coulomb interaction. The acoustic mode with lower phason velocity v_ϕ dominates the spectral weight at high temperatures ($T\ge 0.3T_c$) while the optical mode does at low temperatures. The predicted phason velocity is confirmed by recent neutron-scattering data[11] from a single crystal of $K_{0.3}MoO_3$. Also the scattering data for the transverse phason velocity appear to be consistent with the present theory. Although the amplitudon has been seen in the CDW of blue bronze $K_{0.3}MoO_3$ in the Raman scattering[16] and in the neutron scattering,[14] the optical mode appears not to be seen in any of these experiments. On the other hand, in these experiments as well as in a recent milliwave conductivity[17] in the CDW in $K_{0.3}MoO_3$ another mode has been identified, the nature of which is still unclear. As to the fluctuation in blue bronze $K_{0.3}MoO_3$ the pressure dependence[18] of T_c appears to suggest the model of imperfect nesting. Unfortunately, however, this is not the whole story. Contrary to the expectation of the model with imperfect nesting, the ratio Δ_a/T_c also decreases with pressure.[17] Perhaps a small admixture of the fluctuation effect may be required but certainly more work is desirable on this question.

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APPENDIX A: DERIVATION OF EQ. (3)

In the presence of the long-range Coulomb interaction $\langle[\Delta_2,\Delta_2]\rangle$ is evaluated as[12]

$$\langle[\Delta_2,\Delta_2]\rangle=\langle[\Delta_2,\Delta_2]\rangle_0-\frac{4\pi e^2}{q^2}\frac{\langle[\Delta_2,n]\rangle_0\langle[n,\Delta_2]\rangle_0}{1+\frac{4\pi e^2}{q^2}\langle[n,n]\rangle_0}\ , \tag{A1}$$

where n is the fluctuation of the electron density and $\langle\ \rangle_0$ is the retarded product in the absence of the Coulomb interaction. Then inserting corresponding retarded products, we obtain

$$\langle[\Delta_2,\Delta_2]\rangle=N_0\left\{\frac{1}{\lambda}+\frac{f}{(2\Delta)^2}\left[\omega^2-\zeta^2-\omega_p^2 f\left[1-\frac{\omega_p^2(1-f)}{\omega^2-\zeta^2}\right]^{-1}\right]\right\}\ , \tag{A2}$$

where N_0 is the electron density of states at the Fermi surface per spin and $\lambda=g^2N_0$.

APPENDIX B: OPTICAL MODE FOR AN ARBITRARY q

At $T=0$ K the analysis given in Sec. III B is easily extended for an arbitrary **q**. Now making use of

$$\frac{1}{2\pi}\int_0^{2\pi}d\chi\frac{\zeta^2}{\zeta^2-\omega^2}(1-f)\cong\frac{2}{3}\frac{\zeta_\parallel^2+\zeta_\perp^2}{(2\Delta)^2}\left[1-\frac{4}{5}\left[\frac{\zeta_\parallel^4+6\zeta_\parallel^2\zeta_\perp^2+\frac{3}{2}\zeta_\perp^4}{\zeta_\parallel^2+\zeta_\perp^2}-\omega^2\right](2\Delta)^{-2}+\cdots\right]\ , \tag{B1}$$

$$\frac{1}{2\pi}\int_0^{2\pi}d\chi\,\zeta f=\zeta_{\zeta_\parallel}[1-\tfrac{2}{3}(\zeta_\parallel^2+3\zeta_\perp^2-\omega^2)(2\Delta)^{-2}+\cdots]\ , \tag{B2}$$

where

$$\zeta=\zeta_\parallel+\sqrt{2}\zeta_\perp\cos\chi\ , \tag{B3}$$

and we made use of Eq. (12) in the text, the optical mode is now given by

$$0=\frac{m^*}{m}\omega^2-\langle\zeta^2\rangle+\langle\zeta f\rangle^2\langle f\rangle^{-1}\left\langle\frac{\zeta^2(1-f)}{\omega^2-\zeta^2}\right\rangle^{-1}\ , \tag{B4}$$

where $\langle\ \rangle$ means the average over χ.

Substituting (B1) and (B2) into (B4), we obtain

$$\omega_{\text{op}}^2=\tfrac{3}{2}\lambda\omega_Q^2\zeta_\parallel^2(\zeta_\parallel^2+\zeta_\perp^2)^{-1}+\frac{1}{5}\frac{m}{m^*}(\zeta_\parallel^6+11\zeta_\parallel^4\zeta_\perp^2-6\zeta_\parallel^2\zeta_\perp^4+5\zeta_\perp^6)(\zeta_\parallel^2+\zeta_\perp^2)^{-2}\ . \tag{B5}$$

The energy gap in the optical mode depends strongly on the direction of **q** and vanishes completely when **q**⊥**a**.

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