PHYSICAL REVIEW A VOLUME 18, NUMBER 6 DECEMBER 1978

Backflow effect in classical systems

P. K. Kahol and D. K. Chaturvedi
Department of Physics, Panjab University, Chandigarh-160014, India
(Received 31 January 1978)

The effect of backflow of quasiparticles on the dynamics of classical systems is investigated using the mean-field expression for the density response function proposed recently by Aldrich *et al.* It is observed that the overall results for classical liquids (e.g., liquid argon and rubidium) are qualitatively of the same nature as obtained from earlier mean-field expressions, whereas qualitative and quantitative improvement is achieved for one-component classical plasma.

I. INTRODUCTION

In the conventional mean-field theories[1,2] of the density response function a very important effect known as the backflow of quasiparticles was completely neglected. The backflow effect manifests itself through the vector polarization potential and introduces a frequency and wave-vector-dependent term in the generalized mean-field potential. Aldrich *et al.*[3] incorporated this effect in the density response function and explained theoretically the elementary excitations in ^{4}He and the zero sound in liquid ^{3}He. Very recently, Gupta and Singwi[4] observed that the basic features of the experimental[5] dynamic structure factor of electrons at metallic densities can not be explained through the existing mean-field theories, whatever way one chooses the local-field corrections and the screened response function. However, with the inclusion of the backflow term in the density response function they found an overall good agreement with observed $S(q,\omega)$ in the wave-vector region $0<q<1.5\,q_F$ (q_F is the Fermi wave vector). In view of the above significant effect of backflow in quantum systems, we feel that it would be worthwhile to examine its effect in classical systems also. The systems we have examined are the classical liquids and the classical one-component plasma.

A considerable amount of experimental and theoretical work has already been done in classical liquids. In liquid argon, experiments do not indicate any evidence of well-defined collective excitations, and the mean-field theories[1] are fairly successful in explaining the data. On the other hand, the inelastic neutron scattering[6] and molecular dynamics[7] (MD) experiments in liquid rubidium have unambiguously demonstrated the existence of propagating sound modes up to $q<1.25$ Å^{-1} and the persistence of structure around $q=2$ Å^{-1}. These features could not be accounted for through the earlier mean-field theories. Since the density response function incorporating the backflow effect could explain the elementary excitations in ^{4}He, zero sound in ^{3}He, and collective excitations in electron liquids, one may expect that it might also explain the data on liquid rubidium.

Extensive MD calculations of time-dependent correlation functions in one-component plasma (OCP) have been performed recently by Hansen *et al.*[8] over a wide range of thermodynamic states characterized by the dimensionless parameter

$$\Gamma = e^2/ak_BT \ ,$$

where a is the ion-sphere radius, defined in terms of the number density n as

$$a = (3/4\pi n)^{1/3} \ .$$

In their analysis Hansen *et al.* used various theories of the mean-field type and found poor agreement with MD results. In this paper we shall calculate the spectral functions by incorporating the backflow term in the density response function and thus examine the effect of backflow on the dynamics of OCP.

The outline of the paper is as follows. In Sec. II we give the mathematical formulation and the sum rules used in the calculations. Numerical results on the classical liquids and the classical OCP are discussed in Sec. III. Section IV contains the conclusions of our paper.

II. MATHEMATICAL FORMULATION

A. General definitions

In the polarization potential model, the expression for the density response function $\chi(q,\omega)$ can be written as[3]

$$\chi(q,\omega) = \frac{\chi_{sc}(q,\omega)}{1-[\psi_1(q)+\omega^2\psi_2(q)/q^2]\chi_{sc}(q,\omega)} \ , \qquad (1)$$

where $\chi_{sc}(q,\omega)$ is the response of the system to the external field plus the induced polarization potentials. The latter are defined in terms of the induced

particle ($\langle\langle \rho(q,\omega)\rangle\rangle$) and current density fluctuations ($\langle\langle \vec{j}(q,\omega)\rangle\rangle$) through the relations

$$\phi_{\rm pol}(q,\omega)=\psi_1(q)\langle\langle\rho(q,\omega)\rangle\rangle\,, \tag{2}$$

$$\vec{A}(q,\omega)=\psi_2(q)\langle\langle\vec{j}(q,\omega)\rangle\rangle\,. \tag{3}$$

The functions $\psi_1(q)$, $\psi_2(q)$, and $\chi_{sc}(q,\omega)$ are not known *a priori*. But $\psi_1(q)$ and $\psi_2(q)$ can be calculated with the help of moment sum rules and the form of $\chi_{sc}(q,\omega)$ can be chosen from the existing knowledge.

We consider $\chi(q,\omega)$ to be analytic in the upper half plane so that we may write

$$\chi(q,\omega)=\int_{-\infty}^{\infty}\frac{d\omega'}{\pi}\,\frac{\chi''(q,\omega)}{\omega'-\omega-i\eta}\,, \tag{4}$$

where $\eta=0^+$ and $\chi''(q,\omega)$ denotes the imaginary part of $\chi(q,\omega)$. $\chi''(q,\omega)$ is related to the real part of the density response function $\chi'(q,\omega)$ through the Kramers-Kronig dispersion relation, and to $S(q,\omega)$ through the fluctuation-dissipation theorem, which in the classical limit can be written as

$$S(q,\omega)=-(k_BT/n\pi)\chi''(q,\omega)/\omega\,. \tag{5}$$

B. Sum rules

Using Eq. (5) the large-ω expansion of $\chi(q,\omega)$ in Eq. (4) can be written in terms of the even moments ($\langle\langle\omega^{2l}\rangle\rangle$) of $S(q,\omega)$ as

$$\chi(q,\omega)=\frac{n}{k_BT}\sum_{l=1}^{\infty}\frac{\langle\omega^{2l}\rangle}{\omega^{2l}}\,. \tag{6}$$

Similarly, the screened response function can be expressed as

$$\chi_{sc}(q,\omega)=\sum_{l=1}^{\infty}\langle\omega_{sc}^{2l-1}\rangle/\omega^{2l}\,, \tag{7}$$

where we have defined

$$\langle\omega_{sc}^{2l-1}\rangle=-\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\,\omega^{2l-1}\chi_{sc}''(q,\omega)\,. \tag{8}$$

Now taking the large-ω expansion of Eq. (1) and using (7), we compare the coefficients of various powers of ω in (6) to obtain the following expressions for the second, fourth, and sixth moments of $S(q,\omega)$:

$$\langle\omega^2\rangle=[k_BT/nA(q)]\langle\omega_{sc}^1\rangle\,, \tag{9}$$

$$\langle\omega^4\rangle=\frac{k_BT}{nA(q)}\left[\langle\omega_{sc}^3\rangle+\frac{\langle\omega_{sc}^1\rangle}{A(q)}\left(\psi_1(q)\chi\langle\omega_{sc}^1\rangle+\frac{\psi_2(q)}{q^2}\langle\omega_{sc}^3\rangle\right)\right], \tag{10}$$

$$\langle\omega^6\rangle=\frac{k_BT}{nA(q)}\left\{\langle\omega_{sc}^5\rangle\left[1+\frac{\psi_2(q)}{q^2}\,\frac{\langle\omega_{sc}^1\rangle}{A(q)}\right]+\frac{\langle\omega_{sc}^3\rangle}{A(q)}\left[2\psi_1(q)\langle\omega_{sc}^1\rangle-\frac{\psi_2(q)}{q^2}\left(-\langle\omega_{sc}^3\rangle+2\psi_1(q)\,\frac{\langle\omega_{sc}^1\rangle^2}{A(q)}+\frac{\psi_2(q)}{q^2}\,\frac{\langle\omega_{sc}^3\rangle\langle\omega_{sc}^1\rangle}{A(q)}\right)\right.\right.$$
$$\left.\left.-\psi_1^2(q)\,\frac{\langle\omega_{sc}^1\rangle^3}{A(q)}\right]\right\}, \tag{11}$$

where

$$A(q)=1-[\psi_2(q)/q^2]\langle\omega_{sc}^1\rangle\,. \tag{12}$$

From Eqs. (1), (4), and (5) the zero-moment sum rule gives

$$S(q)=(k_BT/n)[\psi_1(q)-\chi_{sc}^{-1}(q,0)]^{-1}\,. \tag{13}$$

It may be noted that the above expressions are general and are independent of any specific form for the $\chi_{sc}(q,\omega)$. The simplest approximation for the screened response function is to take it equal to that for a noninteracting gas. This choice of $\chi_{sc}(q,\omega)$, however, leads to violation of the second moment. In order to satisfy this moment we make the assumption that $\chi_{sc}(q,\omega)$ is the response of noninteracting quasiparticles with q-dependent "effective mass," defined through the relation

$$m^*(q)=m+n\psi_2(q)\,. \tag{14}$$

With the above assumption the second moment is exactly satisfied and for the zeroth and fourth moment we have

$$S(q)=[(n/k_BT)\psi_1(q)+1]^{-1} \tag{15}$$

and

$$\langle\omega^4\rangle=(q^4k_BT/m^2)[3k_BT+n\psi_1(q)]\,. \tag{16}$$

Clearly the fourth-moment sum rule is violated. In order to incorporate this moment in our model we further assume, in a manner similar to Pathak and Singwi (PS), that there will be left some residual interaction between the quasiparticles which

will have the physical effect of broadening the δ-functions in $[\chi''_{sc}(q,\omega)]_{free}$ with a width $\gamma(q)$. Thus with this choice of $\chi_{sc}(q,\omega)$, relation (10) is exactly satisfied, and from Eqs. (10), (11), and (12) we get

$$\gamma(q) = m[\langle\omega^4\rangle/\langle\omega^2\rangle - \langle\omega^2\rangle(2 + 1/S(q))], \tag{17}$$

$$\psi_1(q) = -(k_B T/2n)[\langle\omega^4\rangle/\langle\omega^2\rangle^2 - 3/S(q)] \tag{18}$$

and

$$\frac{m^*(q)}{m} = \frac{3}{2}\,\frac{[\langle\omega^4\rangle - \langle\omega^2\rangle^2/S(q)]^2}{\langle\omega^2\rangle\langle\omega^6\rangle - \langle\omega^4\rangle^2}\ . \tag{19}$$

As can be seen from Eq. (19), $m^*(q)$ approaches m at large q and we recover the PS model. Another relation of interest which we shall use to fix $m^*(q)$ is

$$S(q,0) = \left[\frac{1}{4\pi}\,\frac{m^*(q)}{m}\left\{\frac{\langle\omega^4\rangle}{\langle\omega^2\rangle} - \frac{\langle\omega^2\rangle}{S(q)}\right\}\right]^{-1}\frac{S^2(q)}{\langle\omega^2\rangle}\ . \tag{20}$$

Thus the central point of the present calculation is that the flexibility introduced in Ref. 1 through $\gamma(q)$ and the flexibility introduced in Ref. 3 through $\psi_2(q)$ has been combined.

III. NUMERICAL RESULTS

A. Classical liquids

In classical liquids we have investigated two systems: (i) liquid rubidium at 319 K and (ii) liquid argon at 76 K. The necessary inputs have been taken from the literature.[6,7,9]

Since in liquid rubidium no experimental or theoretical data are available for the sixth moment, we have fixed $m^*(q)$ from Eq. (20) for $q > 1.25$ Å^{-1}. That the values of $m^*(q)$ are of the same order of magnitude as calculated from Eq. (19) is shown in Fig. 1(b) where we have used approximate values of the sixth moment obtained from our earlier paper.[10] The results are similar in both cases except around $q = 2.0$ Å^{-1} where they differ by a factor of 2. In Fig. 2, we have plotted the results for the dynamic structure factor along with the experimental and the theoretical results of the PS model.[11] For $q < 1.25$ Å^{-1}, with $m^*(q)$ from Eq. (19), we do not get any peak at $\omega = 0$ but the appearance of the peak at finite ω is very distinct. This, more than anything, shows that the basic formulation given in Eq. (1) is probably incapable of giving both the sound peak and the central peak. For $q \simeq 2.0$ Å^{-1}, where the structure in $S(q,\omega)$ still persists, no qualitative improvement over the results of the PS model is found. At large q, the present results almost coincide with the experimental results.

For argon we have chosen the MD data of Rahman[9] for comparison. Since for this choice we have neither very accurate data for the sixth moment nor $S(q,0)$ to fix $m^*(q)$, we have sought optimum values of $m^*(q)$. To justify our numbers for $m^*(q)$, we also calculate it using (i) Eq. (20) and the data of Levesque *et al.*[12] (shown by crosses) and (ii) sixth moments due to Ailawadi *et al.*[13] (shown by solid circles). For all the wave vectors the overall quality of our results is very similar to that of PS but with a difference that now our peak position in the spectral function is in precise agreement with MD results. We demonstrate this

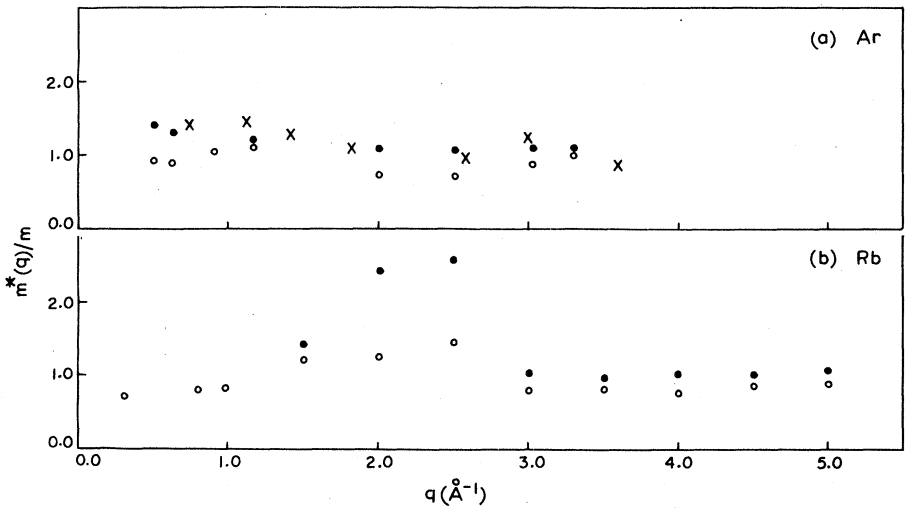

FIG. 1. $m^*(q)$ vs (q). (a) Solid circles denote the results from optimum fit of the data whereas the open circles denote results calculated from the model of Ailawadi *et al.* (b) Solid circles from $S(q,\omega)$ at $\omega = 0$ and open circles from the two-relaxation-time model of Kahol *et al.*

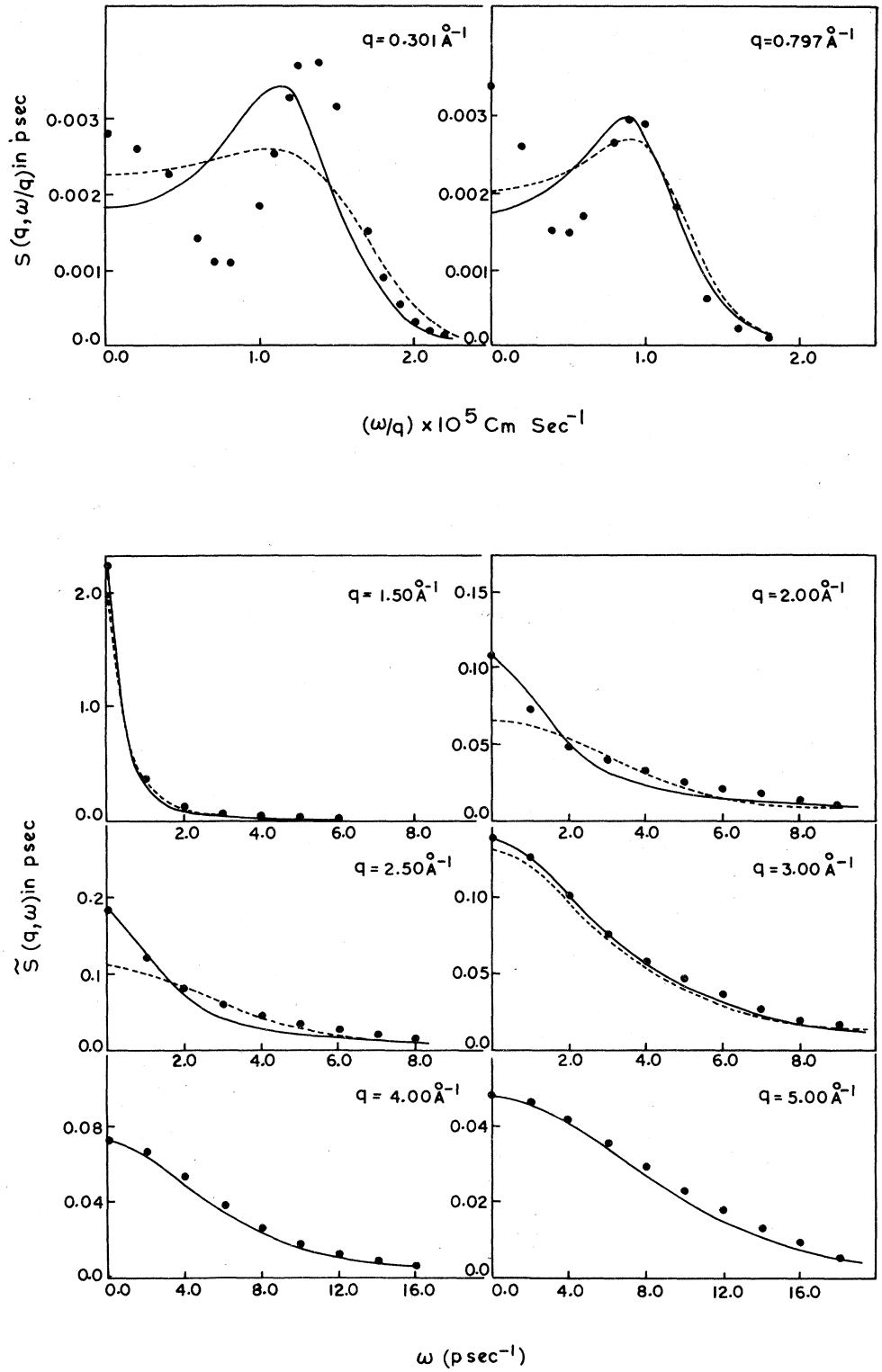

FIG. 2. Scattering function $S(q,\omega/q)$ vs ω/q for $q=0.301$ and 0.797 Å^{-1} along with the molecular dynamics data of Rahman. For $q \geq 1.50$ Å^{-1}, the symmetrized scattering function $\tilde{S}(q,\omega)$ is plotted as a function of ω. The solid circles are the neutron experiment data. The dotted curves are due to Bansal and Pathak.

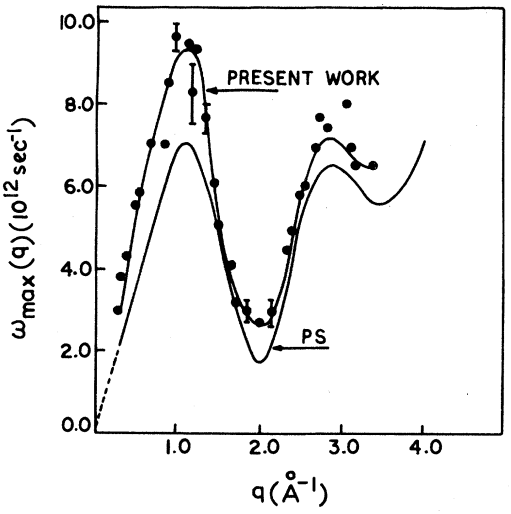

FIG. 3. Dispersion curve $\omega_{max}(q)$ vs q for the longitudinal current fluctuations: solid circles, results of molecular dynamics calculations of Rahman; crosses, experimental results of Sköld and Larsson.

fact in Fig. 3 where the present results almost coincide with MD data.

B. Classical one-component plasma

For OCP, we choose as the unit of time the inverse plasma frequency ω_p^{-1}, defined by

$$\omega_p^2 = 4\pi ne^2/m ,$$

and as the unit of length the ion sphere radius a. In Fig. 4 we have plotted $m^*(q)/m$, obtained from Eq. (19), versus q for three values of Γ, namely, 9.7, 110.4, and 152.4; for $\Gamma = 0.997$ the values of the sixth moment are not available. Since MD results for the zeroth and second moments are not sufficiently accurate, we have taken the values of the Monte Carlo $S(q)$ and the exact second moments. For sixth moment, numerical results[14] have been received which are in good agreement with the fitted values of Hansen *et al.* It is very interesting to see that the wave-vector dependence of $m^*(q)/m$ is quite gentle and approaches unity at large wave vectors. We could not examine the small-q region because of the nonavailability of data for $q < 0.618$. This region is of considerable interest in the sense that at $q = 0$ the value of $m^*(q)/m$ obtained by Gupta and Singwi is 0.88 and that given by Landau theory is about 0.95.

We now discuss systematically the results for the dynamical structure factor of OCP in the various mean-field theories. In the random phase approximation,[8] it is found that $S(q, \omega)$ is too sharp at smaller q's and too flat at larger values of q. If we ascribe the above disagreement to the complete neglect of collisional damping in the theory and incorporate this effect,[1] the improvement in the results (dashed curves in Figs. 5–7) is only marginal. In this context, the results obtained with the present formulation [Eqs. (1) and (17)–(20)] which we display by full lines in Figs. 5–7 are quite significant because, with the inclusion of the backflow term, there is qualitative and quantitative improvement in the results. For example, around $q = 3$, we now get a peak in good agreement with MD results. At large wave vectors the present results almost coincide with MD data. The present formalism is, however, inapplicable in

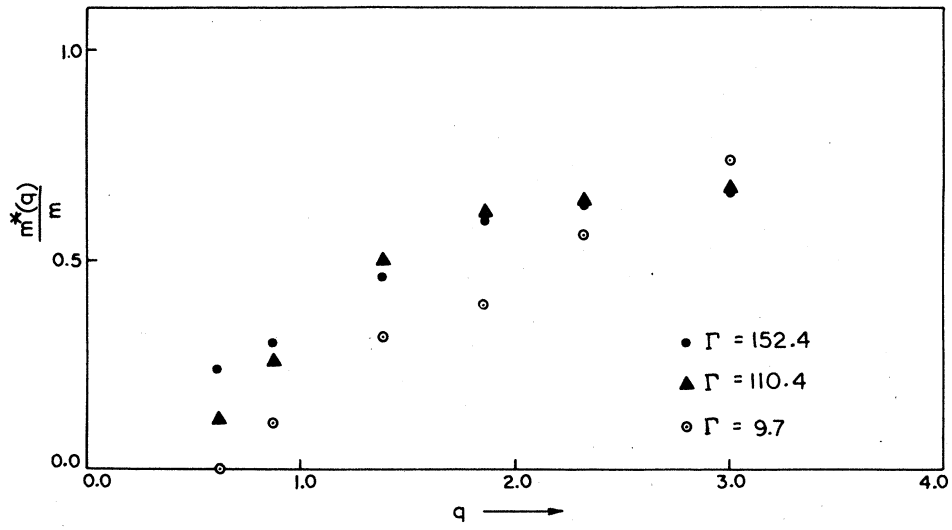

FIG. 4. $m^*(q)$ vs q for different Γ.

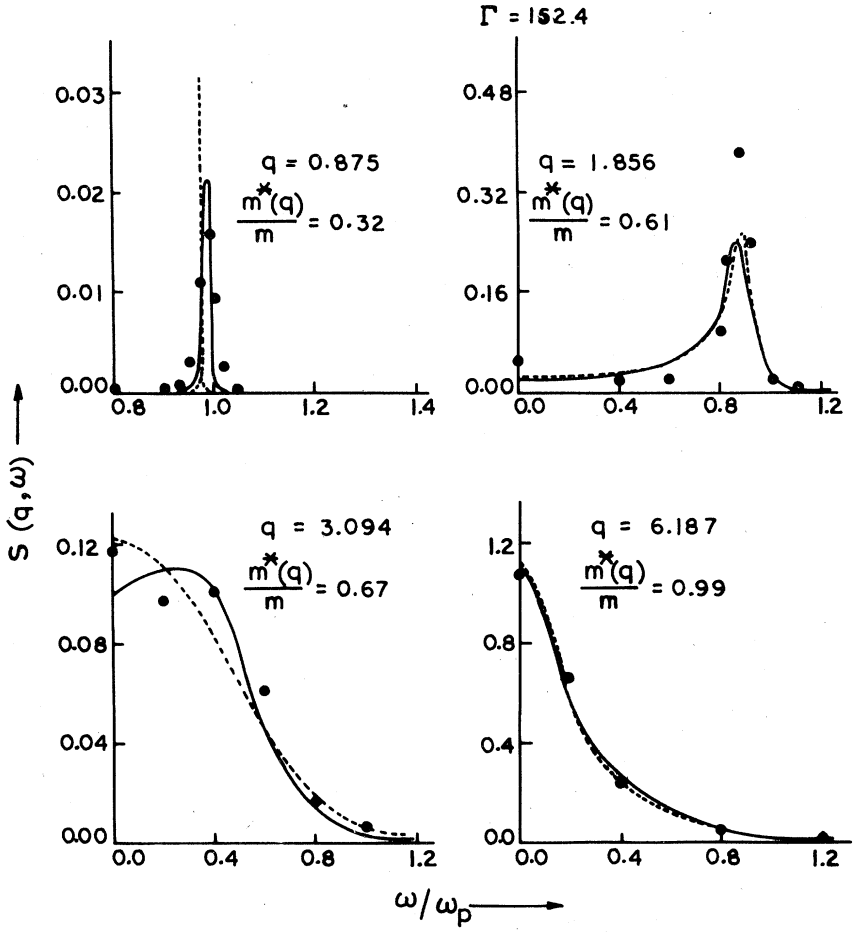

FIG. 5. Scattering function $S(q,\omega)$ vs ω/ω_p. The solid circles denote the MD results and solid and dashed lines are, respectively, the results with and without the backflow term in $\chi(q,\omega)$.

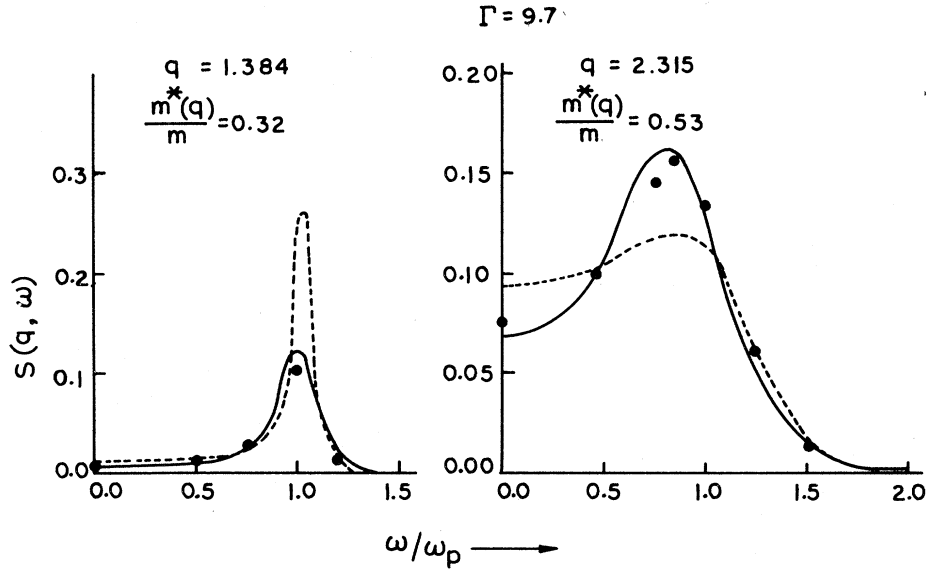

FIG. 6. Same as in Fig. 5.

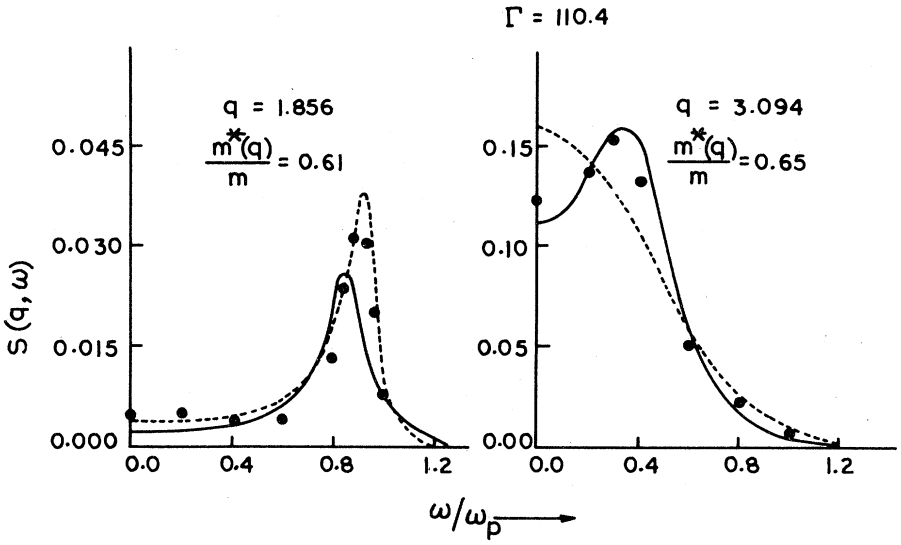

FIG. 7. Same as in Fig. 5.

the small-q region where it gives δ peaks (for $\Gamma = 9.7$ and 110.4).

It may be remarked that the present results which have been obtained without any adjustable parameter are of the same quality as those of Hansen *et al.*[8] obtained by least-squares fit of their MD data.

IV. CONCLUSIONS

In this paper we have investigated the effect of backflow on the dynamics of classical liquids and classical OCP. We find that the magnitude of $m^*(q)/m$ is slightly greater than unity for classical liquids whereas it is always less than unity for classical OCP—gently approaching unity at large wave vectors. This can be understood in terms of the physical properties of the systems—like the interaction potential and the mass of the interacting particle. In classical liquids, the particles interact via weak attractive forces for distances greater than the hard-core radius. The surroundings, therefore, act as a drag on the motion of the "blue" particle and hence the mass of the quasiparticle becomes greater than the bare mass. However, as the particles are massive and the attractive forces weak, the surroundings have only small observable effect. But in OCP, the repulsive nature of the long-range Coulomb potential helps the surroundings to push forward the blue ion and therefore the mass of the quasiparticle is less than the bare mass. Furthermore, since the ionic mass is small and the Coulomb forces are relatively strong, we observe appreciable effect of backflow on the dynamics of OCP. That is why the backflow effect is also important in liquid ^{3}He, ^{4}He, and the electron gas.

The present formalism is incapable of explaining the propagating sound waves and the persistance of structure around $q = 2$ Å^{-1} in liquid rubidum. For both liquid rubidium and liquid argon, the quality of the present results is not much better than that of the PS model. But in OCP there is a qualitative as well as quantitative improvement in the results. In summary, we may conclude that the effect of backflow on the dynamics of classical liquids is small but it is appreciable in systems like classical OCP, electron gas, ^{3}He, and ^{4}He.

ACKNOWLEDGMENTS

We wish to thank Professor K. S. Singwi for suggesting the problem and Professor K. N. Pathak for his comments. One of us (P.K.K.) acknowledges financial assistance from the Council of Scientific and Industrial Research, New Delhi, India.

[1]K. N. Pathak and K. S. Singwi, Phys. Rev. A 2, 2427 (1970).
[2]P. Vashishta and K. S. Singwi, Phys. Rev. B 6, 875 (1972).
[3]C. H. Aldrich III, C. J. Pethick, and D. Pines, Phys. Rev. Lett. 37, 845 (1976); C. H. Aldrich III, Ph.D. thesis (University of Illinois, 1974) (unpublished).
[4]A. K. Gupta and K. S. Singwi (private communication).

[5]P. E. Batson, C. H. Chen, and J. Silcox, Phys. Rev. Lett. 37, 937 (1976).
[6]J. R. D. Copley and J. M. Rowe, Phys. Rev. Lett. 32, 49 (1976).
[7]A. Rahman, Phys. Rev. Lett. 32, 53 (1974); Phys. Rev. A 9, 1667 (1974).
[8]J. P. Hansen, I. R. McDonald, and E. L. Pollock, Phys. Rev. A 11, 1025 (1975).
[9]A. Rahman, Phys. Rev. Lett. 19, 420 (1967); *Neutron Inelastic Scattering* (IAEA, Vienna, 1968), Vol. 1, p. 561.
[10]P. K. Kahol, R. Bansal, and K. N. Pathak, Phys. Rev. A 14, 408 (1976).
[11]R. Bansal and K. N. Pathak, Phys. Rev. A 11, 1450 (1975).
[12]D. Levesque, L. Verlet, and J. Kurkijarvi, Phys. Rev. A 7, 1690 (1973).
[13]N. K. Ailawadi, A. Rahman, and R. Zwanzig, Phys. Rev. A 4, 1616 (1971).
[14]H. B. Singh (private communication).