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COLLECTIVE EXCITATIONS IN A SYSTEM OF PERMANENT ELECTRIC DIPOLES*

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It is argued that for a system of permanent dipoles no collective excitations associated with the charge-density fluctuations exist.

Most assemblies of electric charges such as the electron gas[1] and van der Waals[2] solids are known to exhibit collective motions associated with self-sustained oscillations of the charge-density fluctuations. The purpose of this Letter is to show that a system very closely related to the above, namely a system of permanent electric dipole moments, does not exhibit the above-mentioned collective excitations.

It is well known that the collective motion of system of charges can be studied through the dielectric-function formalism.[3] The dielectric function is the response of the system to an external-probe charge density $\rho_{\text{ext}}(\vec{q},\omega)$ and it has the form

$$\epsilon(\vec{q},\omega)=1-\frac{\langle\rho(\vec{q},\omega)\rangle}{\langle\rho(\vec{q},\omega)\rangle+\rho_{\text{ext}}(\vec{q},\omega)},\tag{1}$$

where $\langle\rho(\vec{q},\omega)\rangle$ is the Fourier transform of the average charge-density fluctuation induced in the system by the external probe.

Self-sustained oscillations occur for ω's satisfying the equation[3]

$$\epsilon(\vec{q},\omega)=0.\tag{2}$$

In case Eq. (2) has no roots for real ω, it might have solutions for complex ω. These roots will lie on the lower half of the complex ω plane, on the second sheet of the Riemannian surface, and we can still think of collective motion (which is now damped) if the damping is small.[4]

The expression for the dielectric function in the linear-response theory is

$$\epsilon(\vec{q},\omega)=1+\frac{4\pi\alpha_0(\vec{q},\omega)}{1-4\pi\alpha_0(\vec{q},\omega)[1-\gamma(\vec{q},\omega)]}.\tag{3}$$

Here $\gamma(\vec{q},\omega)$ takes into account the local field corrections. It is unity in the usual random phase approximation, where the local field is identified with the macroscopic field (i.e., no local-field corrections). $\alpha_0(\vec{q},\omega)$ is the free polarizability, which is given by[5]

$$\alpha_0(\vec{q},\omega)=-(\Omega/q^2)\int_0^\infty dt\,\exp(i\omega t-0^+t)\langle i[\rho(-\vec{q}),\rho(\vec{q},t)]\rangle.\tag{4}$$

Self-sustained oscillations occur when $\epsilon=0$, i.e., when

$$1+\gamma(\vec{q},\omega)4\pi\alpha_0(\vec{q},\omega)=0.\tag{5}$$

In the case of a liquid composed of molecules with permanent electric dipoles, the local field can be calculated by considering a dipole inside a spherical cavity immersed in the dielectric, acted on by a time-dependent external field.[6] The local field is calculated from elementary electromagnetism theory[7] (for $|\vec{q}|\to 0$):

$$\vec{E}_l(t)=\vec{E}_0(t)-\{2\epsilon(\vec{q},\omega)/[2\epsilon(\vec{q},\omega)+1]\}4\pi\vec{P}(t)+(8\pi N/3\Omega)[(\epsilon_0-1)/(2\epsilon_0+1)]\vec{\mu}(t),\tag{6}$$

where the first two terms on the right-hand side represent the cavity field $[\epsilon(\vec{q},\omega)$ is the dielectric function$]$ and the third represents the reaction field which follows $\vec{\mu}(t)$ and therefore uses the static ϵ_0 in its coefficient. As the reaction field is parallel to the dipole moment, it cannot produce any orientational effect upon it.[8] It leads to a constant term in the Hamiltonian of the system which can be neglected.

Equation (6) determines $\gamma(\vec{q},\omega)=2\epsilon(\vec{q},\omega)/[2\epsilon(\vec{q},\omega)+1]$. The dielectric function for a system of permanent dipoles is therefore

$$\epsilon(\vec{q},\omega)=1+\frac{4\pi\alpha_0(\vec{q},\omega)}{1-4\pi\alpha_0(\vec{q},\omega)/[2\epsilon(\vec{q},\omega)+1]}.\tag{7}$$

This equation has no solution for $\epsilon = 0$. This can be seen by the absence of poles of $\alpha_0(\vec{q}, \omega)$ for a dipolar system. It can also be noticed that, by putting $\epsilon = 0$ on the right-hand side, Eq. (7) reduces to the dielectric function of the noninteracting system, which cannot exhibit any collective elementary excitation. Hence, in this approximation no collective excitation associated with the longitudinal polarization charge exists.

It is important to notice that any static model, in which γ is taken to be frequency independent, would indeed lead to collective motions; i.e., there would be a solution of $\epsilon(\vec{q}, \omega) = 0$. A suitable static approximation for the dipolar system would be to take $\gamma = 2\epsilon_0/(2\epsilon_0 + 1)$, which leads to the correct limit as $\omega \to 0$.[9]

It is the more realistic dynamical approximation that is responsible for washing out the collective excitations of the system. It seems clear that although the static approximation works well for the electron gas,[10,11] dynamical local-field effects have always to be taken into account in dealing with assemblies of permanent electric dipoles in particular and, probably, in dealing with systems of localized charges in general.

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[9] Previously (Ref. 5), by using the static approximation, two contradictory expressions for the frequency of the collective oscillations were obtained, one from the dielectric-function method and the other by the equation-of-motion method. The discrepancy can be eliminated if a consistent use of Kubo's linear-response theory is applied in both approaches, and the result is that $\omega_{p1}^2 = \omega_0^2(1 + k_BT/3I\omega_0^2) + O((k_BT/I\omega_0)^2)$, with $\omega_0^2 = 8\pi N\mu^2/3\Omega I$.

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