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Light-scattering study of helium-xenon gas mixtures: Slow sound

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We present light-scattering experiments on various He+Xe gas mixtures. Our results give evidence that the sound mode for large wave vectors propagates with a velocity which is of the order of the adiabatic sound velocity of pure xenon.

Recently, Montfrooij *et al.* published the first experimental results concerning fast sound.[1] In their Letter Montfrooij *et al.*[1] remarked that the damping of the sound mode we have measured by light scattering in He+Xe gas mixtures[2] might be related to the manifestation of a slow sound wave which is supported by the heavy particles. The light-scattering spectrum

$$I(k,\omega)\cong a_i^2x_iS_{ii}(k,\omega)+2a_ia_j(x_ix_j)^{1/2}S_{ij}(k,\omega)+a_j^2x_jS_{jj}(k,\omega) \qquad (1)$$

depends on the partial dynamic structure factors $S_{ij}(k,\omega)$, the mole fraction x_i, and the polarizability a_i of component i. In the case of He+Xe gas mixtures, light-scattering experiments probe only the dynamics of the fluctuations in the xenon density, since the polarizability of xenon is about 20 times larger than the polarizability of helium[3-5] We show here that the hypothesis of Montfrooij *et al.*[1] is valid.

We have performed density-dependent light-scattering experiments on various He+Xe gas mixtures. Here we will give a brief account of these experiments.[4] Our experimental setup has a 90° scattering geometry. An argon-ion laser operating single mode at 514.5 nm is used as a light source and the scattered light is analyzed using a Fabry-Perot equipped with flat plates. The gas mixtures are prepared at room temperature, and are allowed to equilibrate for a few days. Composition and other relevant thermodynamic parameters can be calculated from the measured pressures together with an equation of state up to the third virial coefficient.[2] During one series of density-dependent experiments the composition of the mixture is kept constant. In this manner we are able to study the light-scattering spectrum $I(k,\omega)$ for a large domain of reduced wave vectors kl_{He}. Here k and l_{He} represent the wave vector and the mean free path of a helium particle, respectively:[6]

$$l_{\mathrm{He}}=\{\pi n[x_{\mathrm{Xe}}\sigma_{\mathrm{XeHe}}^2(m_{\mathrm{He}}/m_{\mathrm{red}})^{1/2}+\sqrt{2}x_{\mathrm{He}}\sigma_{\mathrm{He}}^2]\}^{-1}. \qquad (2)$$

In Eq. (2) we have $\sigma_{\mathrm{Xe}}=0.3963$ nm, $\sigma_{\mathrm{He}}=0.263$ nm (Ref. 7), and $\sigma_{\mathrm{XeHe}}=(\sigma_{\mathrm{Xe}}+\sigma_{\mathrm{He}})/2$, while n denotes the number density, and m_{red} the reduced mass: $m_{\mathrm{red}}=m_{\mathrm{Xe}}m_{\mathrm{He}}/(m_{\mathrm{Xe}}+m_{\mathrm{He}})$.

For the mixtures with $x_{\mathrm{Xe}}=0.61$ and 0.45 the Brillouin lines are clearly visible in $I(k,\omega)$ in the range $0<kl_{\mathrm{He}}<0.8$, while for $x_{\mathrm{Xe}}=0.22$ the light-scattering spectrum becomes featureless as kl_{He} exceeds 0.15. The data concerning the sound propagation frequency are extracted from our experiments by fitting all the light-scattering spectra with

$$\pi I(k,\omega)=\frac{A_Dz_D}{z_D^2+\omega^2}+\frac{A_s'z_s'+A_s''(\omega\pm z_s'')}{z_s'^2+(\omega\pm z_s'')^2} \qquad (3)$$

in such a manner that the frequency sum rule[8]

$$2z_s''A_s''=z_DA_D+2z_s'A_s' \qquad (4)$$

is fulfilled and then convoluted with the experimentally obtained instrumental profile which was recorded simultaneously with the light-scattering spectrum. In Eqs. (3) and (4) we have used A_D, A_s', and A_s'' to represent the amplitudes of the Rayleigh line, the Brillouin lines, and the asymmetric contribution to the Brillouin lines, while z_D, z_s', and z_s'' represent the width of the Rayleigh line and the attenuation and the propagation frequency of the sound mode, respectively. On the basis of our calculations[6] it can be argued that the representation of the Rayleigh line by a single Lorentzian is a proper one.

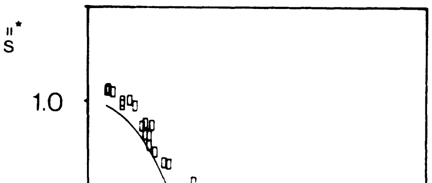

FIG. 1. The propagation frequency of the sound mode reduced with respect to c_sk as a function of the reduced wave vector. The solid line represents the result of a density-dependent hydrodynamic calculation. Thermodynamic circumstances: $T=294$ K, $x_{\mathrm{Xe}}=0.45$.

In Fig. 1 we show for the He+Xe mixture with $x_{Xe}=0.45$ the reduced propagation frequency $z_s''^* = z_s''/c_s k$ as a function of the reduced wave vector kl_{He}. Here, c_s represents the adiabatic sound velocity of the mixture as calculated with the equation of state: $c_s(x_{Xe}=0.45) \cong 261$ ms^{-1}. Also shown in Fig. 1 are the results of a hydrodynamic calculation similar to the one published in Ref. 6. The hydrodynamic theory describes our experimental results well up to $kl_{He} \cong 0.2$, while for the reduced wave vectors $kl_{He} > 0.3$ the propagation frequency of the sound mode tends to the value of $c_s k$ of pure xenon. We have observed similar behavior for He+Xe gas mixtures with $x_{Xe}=0.61$ and 0.22. This is illustrated in Fig. 2 where we show the composition dependence of the observed value of the "sound velocity" z_s''/k in the limit of large kl_{He}. We compare these values with the calculated value of the adiabatic sound velocity of the pure heavy component of the mixture ($x_{Xe}=1$), represented by the solid line, and with the large wave-vector limit of the simplified hydrodynamic theory of Gornall and Wang[9] and Lekkerkerker and Boon[10] (represented by the dashed line; see also Ref. 6). Our experimental results for large kl_{He} indicate the existence of a slow sound wave which propagates with the sound velocity of pure xenon. The equivalent fast sound mode (a wave propagating with the speed of the light component) cannot be detected in this light-scattering experiment since the difference between the polarizabilities of xenon and helium is too large.[4,5]

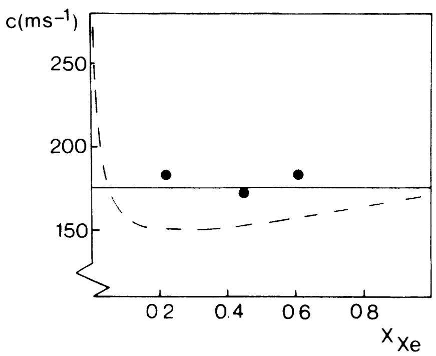

FIG. 2. The "sound velocity" as obtained from light-scattering spectra in the large kl_{He} limit. The solid line indicates the value of the adiabatic sound velocity of pure xenon ($x_{Xe}=1$). The dashed line represents the large wave-vector limit of the simplified hydrodynamic model of Refs. 9 and 10.

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