UvA-DARE (Digital Academic Repository)

Light-Scattering Study of Helium-Xenon Gas-Mixtures - Slow Sound

Wegdam, G.H.; Schaink, H.M.

DOI

[10.1103/PhysRevA.41.3419](https://doi.org/10.1103/PhysRevA.41.3419)

Publication date

1990

Published in

Physical Review A

[Link to publication](#)

Citation for published version (APA):

Wegdam, G. H., & Schaink, H. M. (1990). Light-Scattering Study of Helium-Xenon Gas-Mixtures - Slow Sound. *Physical Review A*, 41, 3419-3420.
<https://doi.org/10.1103/PhysRevA.41.3419>

General rights

It is not permitted to download or to forward/distribute the text or part of it without the consent of the author(s) and/or copyright holder(s), other than for strictly personal, individual use, unless the work is under an open content license (like Creative Commons).

Disclaimer/Complaints regulations

If you believe that digital publication of certain material infringes any of your rights or (privacy) interests, please let the Library know, stating your reasons. In case of a legitimate complaint, the Library will make the material inaccessible and/or remove it from the website. Please Ask the Library: <https://uba.uva.nl/en/contact>, or a letter to: Library of the University of Amsterdam, Secretariat, Singel 425, 1012 WP Amsterdam, The Netherlands. You will be contacted as soon as possible.

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

G. H. Wegdam and H. M. Schaink

Laboratory for Physical Chemistry, University of Amsterdam, Nieuwe Achtergracht 127,
NL-1018 WS, Amsterdam, The Netherlands

(Received 25 September 1989)

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 mixtures² might be related to the manifestation of a slow sound wave which is supported by the heavy particles. The light-scattering spectrum

$$I(k, \omega) \cong \alpha_i^2 x_i S_{ii}(k, \omega) + 2\alpha_i \alpha_j (x_i x_j)^{1/2} S_{ij}(k, \omega) + \alpha_j^2 x_j S_{jj}(k, \omega) \quad (1)$$

depends on the partial dynamic structure factors $S_{ij}(k, \omega)$, the mole fraction x_i , and the polarizability α_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 helium³⁻⁵. 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

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

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

For the mixtures with $x_{\text{Xe}} = 0.61$ and 0.45 the Brillouin lines are clearly visible in $I(k, \omega)$ in the range $0 < kl_{\text{He}} < 0.8$, while for $x_{\text{Xe}} = 0.22$ 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

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

in such a manner that the frequency sum rule⁸

$$2z_s'' A_s'' = z_D A_D + 2z_s' A_s' \quad (4)$$

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 calculations⁶ 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 $c_s k$ 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, $x_{\text{Xe}} = 0.45$.

In Fig. 1 we show for the He+Xe mixture with $x_{\text{Xe}}=0.45$ 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: $c_s(x_{\text{Xe}}=0.45)\cong 261\text{ ms}^{-1}$. 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 $kl_{\text{He}}\cong 0.2$, while for the reduced wave vectors $kl_{\text{He}}>0.3$ 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 $x_{\text{Xe}}=0.61$ 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 ($x_{\text{Xe}}=1$), represented by the solid line, and with the large wave-vector limit of the simplified hydrodynamic theory of Gornall and Wang⁹ and Lekkerkerker and Boon¹⁰ (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 ($x_{\text{Xe}}=1$). The dashed line represents the large wave-vector limit of the simplified hydrodynamic model of Refs. 9 and 10.

We gratefully acknowledge the discussions we have had with I. M. de Schepper and we would like to thank him for sending us Ref. 1 prior to publication. This work is part of the research program of the Foundation for Fundamental Research of Matter (FOM), supported by the Netherlands Foundation for Chemical Research (SON) and was made possible by financial support from the Netherlands Organization for Scientific Research (NWO).

¹W. Montfrooij, P. Westerhuijs, V. O. de Haan, and I. M. de Schepper, Phys. Rev. Lett. **63**, 544 (1989).

²H. M. Schaik and G. H. Wegdam, Physica **141A**, 211 (1987); **160A**, 117 (1989).

³R. C. Burns, C. Graham, and A. R. M. Weller, Mol. Phys. **59**, 41 (1986).

⁴H. M. Schaik, thesis, University of Amsterdam, 1989.

⁵A. Campa and E. G. D. Cohen, Phys. Rev. A **39**, 4909 (1989).

⁶G. H. Wegdam and H. M. Schaik, Mol. Phys. **65**, 531 (1988).

⁷J. O. Hirschfelder, C. F. Curtiss, and R. B. Bird, *Molecular Theory of Gases and Liquids* (Wiley, New York, 1967).

⁸I. M. de Schepper, P. Verkerk, A. A. Van Well, and L. A. de Graaf, Phys. Rev. Lett. **50**, 974 (1983).

⁹W. S. Gornall and C. S. Wang, J. Phys. (Paris) **33**, Suppl. 2-3, C1-51 (1972).

¹⁰H. N. W. Lekkerkerker and J. P. Boon, Phys. Lett. **39A**, 9 (1972).