Possible application of the nuclear γ-resonance technique to the investigation of the elastic properties of liquids

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A calculation is reported of the probability of resonance absorption of γ rays by a colloidal particle in a liquid dispersive medium, assuming that the particle executes harmonic oscillations due to the elastic properties of the medium. An experimental study has been carried out of the probability of resonance absorption of γ rays as a function of the elasticity modulus of the dispersive medium for organosols of β-stannic acid. The experimental and theoretical resonance absorption data are compared.

1. INTRODUCTION

The observation of the Mössbauer effect in liquids[1,2] appears to be an argument in favor of the quasicrystalline model of a liquid.[3,4] However, the appearance of the Mössbauer effect in liquids is restricted to highly viscous liquids and a relatively narrow temperature range. The possibility of using nuclear γ resonance (NGR) as a tool for investigating the viscoelastic properties of liquids are substantially extended when particles with linear size between 50 and 1000 Å are used as the Mössbauer probes instead of the individual atoms. In general, the motion of such a particle can be imagined as a combination of oscillatory, rotational, and diffusion motions. The presence of only diffusion displacements leads to the broadening of the NGR line.[5,6] Oscillatory motions of the colloidal particles was taken into account in our previous paper.[7]

The aim of the present work was to investigate the effect of oscillatory motions of a particle, which are connected with the elastic properties of the liquid, and the probability of γ resonance with a view to using this result to investigate the elastic properties of the liquid. The experimentally observed reduction in the probability of resonance absorption in colloidal solutions with decreasing viscosity[8] was regarded as justification for this formulation of the problem.

2. PROBABILITY OF RESONANCE ABSORPTION BY A COLLOIDAL PARTICLE IN THE CASE OF OSCILLATORY MOTION

Consider a set of small crystalline particles containing Mössbauer nuclei and executing oscillations in a matrix in the form of a viscoelastic liquid. We shall assume that the oscillations of the particles in the liquid are independent of the motion of the Mössbauer nuclei. This is justified if the coupling between the atoms and the particle is much greater than coupling in the liquid. We shall confine our attention to recoilless transitions in the particle, modulated by the oscillations of the particle in the liquid. It can be shown[7] that the absorption spectrum will then contain the unshifted Mössbauer line with a natural width connected with the transfer of the γ-ray momentum to the particle-liquid system as a whole. The intensity of this line is determined by the probability of resonance absorption $f = f_c f_p$, where f_c is the usual Debye-Waller factor for the crystal (in the case of microparticles, it must include corrections for surface effects[9,10]) and f_p is an analog of the Debye-Waller factor which takes into account the oscillations of the particle in the liquid.

In the Debye model, the distribution of the particle oscillation frequencies may be written in the form[7]

$$f_p = \exp\left[-\frac{6RkT}{(\hbar\Omega_1)^2}\frac{1-\alpha}{1-\alpha^3}\right]. \tag{1}$$

In this expression, $R = \varkappa^2\hbar^2/2M$, M is the particle mass, $\varkappa$ is the wave vector of the γ ray, $\alpha = \Omega_2/\Omega_1$, and Ω_1, Ω_2 are the maximum and minimum oscillation frequencies of the particle. The factor $(1-\alpha)/(1-\alpha^3)$ where $\alpha < 1$ has very little effect on the variation in the resonance absorption probability although the frequencies Ω_1 and Ω_2 may depend appreciably on the temperature. We shall assume henceforth that $\alpha \ll 1$ and will not take α into account.

The frequency Ω_1 is determined by the elastic constants of the liquid and the mass of the particle. For a simple model $\Omega_1^2 = ES/Ml$, where E is the elastic modulus of the liquid, S is the cross section of the particle, and l is the thickness of the deformed layer of the liquid during the compression or extension time. The thickness of this layer is obviously equal to a quarter of the wavelength of the sound wave in the liquid. Using the dispersion relation in the form $\Omega = (2\pi\lambda)(E/\rho_l)^{1/2}$ where λ is the wavelength of the sound wave and ρ_l is the density of the liquid, and substituting $l = \lambda/4$, we obtain the following expression for the probability of resonance absorption of the γ ray by the colloidal system:

$$f = f_c \exp\left[-\frac{\pi\varkappa^2 kT\rho_c}{Er\rho_l}\right]; \tag{2}$$

where it is assumed that the particle is a sphere of radius r and ρ_c is the density of the particle.

We shall rewrite (2) in the form

$$f = f_c \exp\left[-\text{const}\cdot\frac{T}{E(T)r}\right]. \tag{2'}$$

According to this expression, the resonance absorption probability for a colloidal particle oscillating in a liquid is determined not only by the Debye-Waller factor for the particle but also by its size and the elastic properties of the liquid. The result is a more rapid variation with temperature.

If we determine f_p experimentally, we can readily calculate the elastic modulus of the liquid containing the particle which exhibits the resonance absorption of γ rays.

3. EXPERIMENT

We have carried out an experimental examination of the dependence of f on the elastic properties of the liquid using the organosols of β-stannic acid in glycerin, epoxy resin ÉD-5, and adhesive BF-2. We first obtained a hydrosol during the peptization of β-stannic acid dried at room temperature, obtained by oxidation of metallic tin

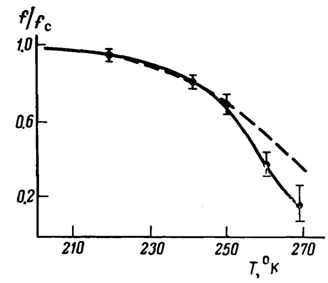

FIG. 1

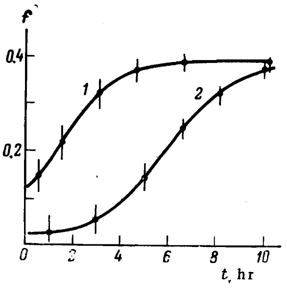

FIG. 2

by nitric acid. The water in the hydrosol was then replaced by glycerin and was evaporated from solution at 140°C over a period of a few hours. The colloidal particles were nearly spherical.

The measurements were carried out on an NGR spectrometer of the electrodynamic type, using constant acceleration and the usual transmission procedure. The colloidal solution was used as the absorber. The γ-ray source was $Sn^{119m}O_2$. To determine the probability of resonance absorption, we used the area under the resonance absorption curve.

The temperature dependence of the probability of resonance absorption of the glycerosol of stannic acid, f, normalized to the probability of resonance absorption of the stannic acid particles, f_c, is shown in Fig. 1 (solid curve). The necessity for normalization was dictated by the variation of f_c in the temperature range under investigation. To determine f_c, the particles were separated from the sol by the addition of acetone followed by drying at room temperature. The broken curve in Fig. 1 shows f/f_c calculated from (2′). The elastic modulus E(T) was determined from the measured velocity of ultrasound in glycerin,[11] extrapolated to higher frequencies. The constant in (2′) was chosen for $f/f_c = 0.8$ and $T = 250°K$. As can be seen, the agreement between the experimental values of f/f_c and those calculated from (2′) is satisfactory if we recall the simplified character of the harmonic oscillator model.

For polymer liquids such as epoxy resin and the BF adhesive, the elasticity may be affected by polymerization at constant temperature. In our investigations, the change in the elasticity of the dispersive medium by polymerization is more convenient because, in this case, f_c is a constant and the change in the probability of resonance absorption of γ rays is completely due to the change in elasticity of the dispersive medium.

Figure 2 shows the probability of resonance absorption of γ rays by the colloidal solution of β-stannic acid in epoxy resin (curve 1) and the BF adhesive (curve 2) as a function of the polymerization time t of the dispersive liquid. The degree of polymerization and, consequently, the elasticity of the dispersive medium increase with time, which leads to a substantial increase in the probability of resonance absorption.

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