Singular vibronic interaction in liquids: manifestation in the optical spectrum of impurity atoms in superfluid helium

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Abstract. The zero phonon line (ZPL) and its phonon-roton wing have been studied both experimentally and theoretically in the optical spectrum of the inner shell transition in the Dy atom in superfluid helium. It is shown that the linear vibronic interaction of impurity atom with long-wave acoustic phonons in the liquid phase is singularly enhanced. As a result, the ZPL of both the superfluid and normal components of liquid helium has a finite width. The temperature dependence of the spectrum is a consequence of the redistribution of the superfluid and normal components of the liquid helium and the temperature dependence of the spectrum of its normal component. Our calculations of the ZPL and its phonon-rotor wing are consistent with the experiment.

1. Introduction

To date, a significant number of experimental and theoretical studies have been conducted on the optical spectra of small molecules in superfluid ^{4}He and ^{3}He droplets (see, for example, book [1], review article [2] and publications [3-7]). In recent years, several experimental studies have been also carried out on the optical spectra of impurity atoms in volume of superfluid ^{4}He [8-15] (see also review article [16]). Therefore, it seems appropriate to consider what differences arise in the optical spectra in the superfluid and in the liquid phase as a whole in comparison with such spectra in crystals and in small droplets. In [17] we recently presented the theory of the zero-phonon line (ZPL) in the optical spectrum of an atom placed in a volume of the superfluid ^{4}He. We have shown that in liquids, due to isochoric conditions (in the macroscopic limit they correspond to the Archimedes Principle), the electronic transition in an atom leads to a finite (albeit microscopic) change in the volume of the entire liquid. There are no such changes in the solid phase. This change in the volume of the liquid is the result of vibronic interaction with long-wave acoustic modes of low, including zero frequency. In solids, such a volume change does not occur, and, consequently, there is no vibronic interaction with acoustic phonons, the frequency of which is close to zero. The same is true for nano-droplets, in which long-wave phonon modes are simply absent. As a result, in liquids, in comparison with solids and nano-droplets, there is a significant increase in the linear vibronic interaction of impurity atoms with low-frequency (long-wave) phonons: in liquids, this interaction became very large with decreasing frequency ω, while in solids and nano-droplets it disappears at low frequencies.

In this communication, we consider the effect of the amplification of the vibronic interaction with long-wave acoustic phonons on the optical spectra of impurity atoms in the superfluid ^{4}He, taking into account the presence of superfluid and normal components in it. We show that this amplification is especially significant for zero-phonon transitions. Indeed, in the solid phase, the probability of these transitions in the case of linear vibronic interaction is finite. However, in the liquid phase, due to the amplification of the vibronic interaction with low-frequency phonons, this probability tends to zero. As a

result, only transitions involving phonons occur in liquids. The latter are more probable the lower the frequency of phonons. As a result, instead of a ZPL of natural width, a broadened ZPL appears in the spectrum [17].

We also show that with increasing temperature, the ZPL in the spectrum of superfluid helium broadens due to an increase in the weight of the normal component, and increase of the width of this component with temperature. Our calculations of the temperature changes of the ZPL of the superfluid [4]He are in good agreement with the results of ZPL measurements [15]. We also demonstrate that the enhanced vibronic interaction with long-wave phonons explains the redistribution of peaks in the single-phonon-rotonic wing of the ZPL, observed in [14], in favour of low-frequency ones.

2. Optical spectrum of impurity center in a liquid

To describe the optical spectrum of an atom in a superfluid He, we use the Lax theory [18] of electronic transitions in optical centres in the case of the linear vibronic interaction

$$V \equiv \hat{H}_2 - \hat{H}_1 = \Omega_0 + \sum_k a_k x_k . \tag{1}$$

Here $\hat{H}_1 = \sum_k \omega_k^2 x_k^2 / 2$ and $\hat{H}_2 = \sum_k \omega_k^2 (x_k + x_{0k})^2$ are the vibrational Hamiltonians of the centre in initial (1) and final (2) electronic states in harmonic approximation, respectively, Ω_0 is the frequency of the electronic transition, $x_{0k} = a_k / \omega_k^2$ is the change of the equilibrium position of the phonon coordinate x_k of the mode k during the transition, ω_k is its frequency, $\hbar = 1$. In Condon approximation, the Fourier transform of the optical spectrum equals to

$$F(t) = \exp\left(i\bar{\Omega}_0 t - \gamma_0 |t| + g(t)\right), \tag{2}$$

where $\bar{\Omega}_0 = \Omega_0 - \Omega_S$ is the frequency of the zero-phonon transition, $\Omega_S = \sum_k \omega_k^2 x_{ok}^2 / 2$ is the Stokes losses, γ_0 is the width of the excited electronic level,

$$g(t) = \alpha \int_0^{\omega_m} d\omega \upsilon_\omega \left((n_\omega + 1)\left(e^{i\omega t} - 1\right) + n_\omega \left(e^{-i\omega t} - 1\right)\right), \tag{3}$$

up to a constant $-S$, is the Fourier transform of the spectrum of the single-phonon transitions [18], $S = \int_0^{\omega_m} d\omega \upsilon_\omega (2n_\omega + 1)$ is the Huang-Rhys factor,

$$\upsilon_\omega = \omega x_{0,\omega}^2 \rho(\omega)/2 \tag{4}$$

is the vibronic interaction for phonon-roton excitation with the frequency ω, $x_{0\omega} \equiv x_{0k_\omega}$, $\rho(\omega)$ is the density of states (DOS), $n_\omega = (e^{\omega/k_B T} - 1)^{-1}$ is the Planck occupancy factor, $\omega_m = 0.55$ THz is maximum frequency of phonons in liquid [4]He.

Let us consider the vibronic interaction υ_ω for the long wave phonons. Taking into account that for small k $\omega = v_0 k$, where $v_0 = 250$ m/s is the velocity of the first sound, we get that in the liquid [4]He in the long-wave limit $\rho(\omega) \approx 0.144 \omega^2$ GHz^{-3}. That is, $\rho(\omega) \propto \omega^2$, as it should be for three-dimensional systems. To find υ_ω for low frequency phonons, we also need to know the dependence $x_{0\omega}$

2

of low frequency modes on ω in a liquid. For this purpose, the isochoric assumption in liquids can be used. According to this assumption, the change in the volume of the liquid due to the electronic transition in the atom should be finite and approximately equal to the change in the volume of the surrounding atoms. In the macroscopic limit, this corresponds to the Archimedes Principle - the total increase in the volume of the liquid and the body immersed in it is equal to the volume of the body.

Assuming that the atom is placed in the centre of a spherical liquid with a radius R, the volume change at the electronic transition in the atom equals $\Delta V = 4\pi R^2 \langle \Delta R \rangle$, where ΔR is the change of the radius of the liquid, $\langle ... \rangle$ is the averaging over the R (we take into account that R can vary at different points on the surface of the sphere at several atomic lengths). In liquids, by virtue of the isochoric assumption, ΔV is finite and for large R do not depend on R. This is the case if $\Delta R \propto R^{-2}$ (see Fig.1).
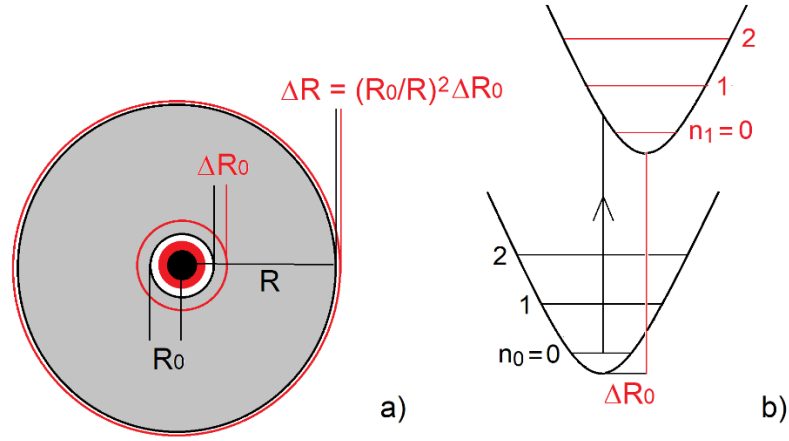

Figure 1. (a) A spherical liquid helium of radius R (grey circle) encapsulating within a cage of radius R_0 an impurity atom (black circle). An increase ΔR_0 of the radius R_0 caused by an electronic transition of the impurity atom which increases its size (red area), is accompanied by an increase ΔR of the external radius, given according to the isochoric conditions (Archimedes Principle in macroscopic limit) by $\Delta R = (R_0/R)^2 \Delta R_0$, and a corresponding increase in the surface area of $8\pi (R_0^2/R)\Delta R_0$. (b) The potential energy curves of the He droplet with respect to the above radial deformation in the ground state (lower parabola) and excited state (upper parabola) of the impurity atom with the respective vibrational states (phonon numbers $n_{0,1} = 0, 1, 2...$). According to the Franck-Condon description, the electronic transition (vertical arrow) occurs on a much faster time scale than that of vibrations and, although starting from a zero-phonon ground state, reaches an excited state with some phonon population, i.e., with non-zero probabilities of $n_1 > 0$ states.

The dependence of $x_{0\omega}$ on ω for low frequency modes can now be found if we take into account that for nondegenerate electronic states of the atom, only $\propto r^{-1}\sin(kr)$ modes contribute to the long-wave vibronic interaction (r is the distance to the origin; another real totally-symmetric solution of the wave equation for the long-wave acoustic modes, $r^{-1}\cos(kr)$, is not suitable while it diverges at $r \to 0$). Therefore, the change in the radius of the liquid due to the electronic transition in the atom can be taken as

$$\langle \Delta R \rangle \propto R^{-1} \left\langle \int_0^{k_m} dk k^2 x_{0k} \sin(kR) \right\rangle \tag{5}$$

3

($k_m = 3.6\,A^{-1}$ is the maxim wave number [19]). Given that $\langle\cos(k_m R)\rangle_0 \simeq 0$, we find that $\Delta R \propto R^{-2}$ and ΔV is finite for $R \to \infty$ if $x_{0k} \propto k^{-2}$, $k \to 0$. Given also that $k \propto \omega$ for small ω, we obtain $x_{0\omega} \propto \omega^{-2}$. Consequently, in liquids, in the low frequency limit

$$\upsilon_\omega \simeq \alpha\omega^{-1}, \qquad (6)$$

where α is a dimensionless parameter of the magnitude of the linear vibronic interaction. In contrast, in solids in the low frequency limit $\upsilon_\omega \sim \omega$ [18,20,21]. Consequently, in liquids, the linear vibronic interaction with low-frequency phonons is amplified by a factor of $\sim \omega_m^2/\omega^2$ compared to this interaction in solids. As a result, this interaction in liquids at $\omega \to 0$ diverges as ω^{-1}. Note that because of this divergency, the Huang-Rhys factor S, which describes the logarithm of the inverse probability of the zero-phonon transitions, also diverges in liquids. This means that in the case of linear vibronic interaction in liquids, there are no zero-phonon transitions, but only transitions involving phonons occur. The latter are more probable the lower the frequency of phonons. As a result, instead of a narrow ZPL, a broadened ZPL appears in the spectrum [17]. In a sense, ZPL in a liquid in the case of linear vibrionic interaction is similar to ZPL in crystals in the case of quadratic vibrionic interaction at $T > 0$: there is also no zero-phonon transition; instead of a ZPL of natural width, a narrow band appears, broadened due to vibronic transitions with the creation and simultaneous destruction of phonons of close frequency (the Raman mechanism of the ZPL broadening) [20].

The dimensionless interaction magnitude parameter α can be estimated using found in [14] increase in the bubble radius at the electronic transition in the impurity $\Delta R_0 \approx 0.15\,\text{Å}$. Presenting the Stokes losses in the form $\Omega_S \approx \bar{\omega}^2 M_4 \delta R^2/2$, where $\bar{\omega}$ is the mean frequency of vibrations of surrounding helium atoms $(\Delta R_0)^2$ is the mean value of $x_{0\omega}^2/M_4$, M_4 is the mass of the helium atom, we get $\alpha \approx (\Delta R_0/A_0)^2$ where $A_0 = \sqrt{\hbar/M_4\bar{\omega}}$ is the mean amplitude of the zero-point vibrations of the helium atoms. Taking for $\bar{\omega} = 0.25$ THz, we get $A_0 \approx 0.75\,\text{Å}$, which gives $\alpha \approx 0.04$.

Note that the enhancement law (6) can also be applied for droplets of large radius R, assuming that the surface tension of the droplet can be neglected. The latter is the case if the contribution $\omega_{ST} = 8\pi\tau R\Delta R$ of the surface tension τ to the energy of electronic transition is small as compared to the ZPL width (see Fig. 1). Taking $\tau \approx 15$ GHz Å^{-2} for superfluid ^{4}He [22], $\Delta R = (R_0/R)^2\Delta R_0$, $R_0 = 5\,\text{Å}$ and $\Delta R_0 \sim 0.15$ Å, we obtain that the surface tension makes a contribution ≥ 1 GHz to the energy of the electronic transition for droplets of radius $R \leq 2\cdot10^3$ Å. For large droplets, the surface tension does not matter.

3. Temperature dependence of the zero-phonon line in superfluid helium

In [15], the temperature dependence of the ZPL in the absorption spectrum of Dy atoms in superfluid helium in the range from 1.4 K to 2.1 K was investigated. The results of measurements are given in Fig. 2 (points). The superfluid liquid ^{4}He has two components: superfluid and normal. The temperature

dependence of the density of normal components can be approximated as $\rho_N \approx (T/T_C)^{6.8}$ [23-25] (see Fig. 2 (left); $T_C = 2.17$ K is the critical temperature); the density of superfluid component equals $\rho_S = 1 - \rho_N$. According to this relation, at 1.4 K $\rho_S = 0.93$ and liquid He is almost entirely superfluid. This component has zero entropy (and zero temperature). Hence, ZPL at 1.4 K basically corresponds to this line for $T = 0$ described by $g(t)_{T=0} \equiv g_0(t) = \alpha \int_0^{\omega_m} d\omega \upsilon_\omega \left(e^{i\omega t} - 1\right)$. Note that Huang-Rhys factor S also diverges at $T = 0$. Consequently, the ZPL in the spectrum of superfluid component has a finite width, and its shape is determined by the vibronic interaction.

To find the ZPL shape, we take into account that the width of the excited level $\gamma_0 \simeq 5\,\text{GHz}$ [15] is small as compared to the width 0.55 THz of the phonon-roton spectrum. Therefore, for the actual values of $t \sim \gamma_0^{-1}$ the exponential in the ω integral for $g_0(t)$ is large. Taking for large $|t|$ $\left(e^{i\omega t} - 1\right)/\omega \cong i\pi \, \text{sgn}(t)\delta(\omega)$, we obtain the following simple equation $g_0(t) = i\pi\alpha \, \text{sgn}(t)$.

In the temperature range $1.4\,\text{K} \geq T \geq 2.1\,\text{K}$ the temperature dependence of the ZPL is essentially determined by the redistribution between the superfluid and normal components. Taking this circumstance into account, the optical spectrum of the centre can be represented in the form $I(\Omega) = \rho_S I_0(\Omega) + \rho_N(T)I_T(\Omega)$, where the spectrum $I_0(\Omega)$ of the superfluid component while $I_T(\Omega)$ is the spectrum of normal component. The Fourier transform of this component is given by equation (2), where $g(t)$ is given by the equation (3). For narrow ZPL as compared to $k_B T$, the main contribution to $g(t)$ comes from small $\omega \ll k_B T$ and large $t \gg (k_B T)^{-1}$. Taking for small ω $n_\omega \simeq k_B T/\omega$, for large $|t|$ $\left(\cos(\omega t) - 1\right)/\omega^2 \cong -\pi|t|\delta(\omega)$ and $\sin(\omega t)/\omega \cong \pi \, \text{sgn}(t)\delta(\omega)$, we obtain $g(t) = -\gamma_T |t| - i\varsigma \, \text{sgn}(t)$, where $\gamma_T = \pi\alpha k_B T$. This gives the ZPL spectrum as the following sum of two lines:

$$I(\Omega) \approx \rho_S \frac{\gamma_0 \cos\varsigma + \Omega \sin\varsigma}{\Omega^2 + \gamma_0^2} + \rho_N \frac{\gamma_N \cos\varsigma + \Omega \sin\varsigma}{\Omega^2 + \gamma_N^2}, \qquad (7)$$

where $\gamma_N = \gamma_0 + \pi\alpha k_B T$ describes the temperature broadening of the ZPL of a normal liquid, $\varsigma = \pi\alpha$ is the asymmetry of ZPL. In the case under consideration $\varsigma \sim 0.1$ and the spectrum consists of two lines of almost Lorentzian shape. The first line $\propto \rho_S$ decreases to zero as the temperature rises to T_C. The intensity and width of the second line increase with temperature, and the increase in width is linear.

In [15], to find the widths of the ZPL, a single-Lorentzian fit of the spectral shape was used. To calculate the width of the ZPL, we also use a single-Lorentzian fitting procedure with the width of the superfluid and normal components $5.15\,\text{GHz}$ and $\gamma_N = 5.15 + 2.5 \cdot T$ GHz, respectively (T in K). The obtained temperature dependence of the ZPL width agrees well with the experiment at $T < 2\,\text{K}$, but at $T \approx 2\,\text{K}$ it gives about 10 percent less value than in the experiment. A possible reason for the discrepancy is the inhomogeneous broadening of the ZPL of normal component stemming from small almost static fluctuations of the bubbles surrounding impurity atoms [26, 27]. Taking the inhomogeneous broadening

in the form $\Gamma\rho_N$ with $\Gamma = 2.5$ GHz, wee obtain the temperature dependence of the ZPL width in good agreement with the experiment (see Fig. 2, right).
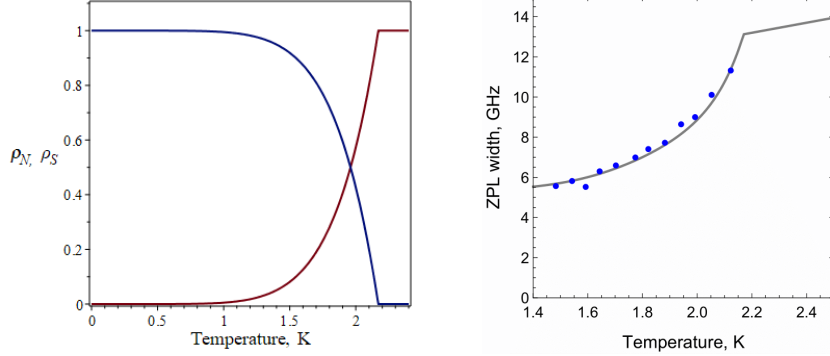

Fig. 2. Temperature dependence of the superfluid (blue) and normal (brown) components of the superfluid helium (left) and width of the ZPL of Dy atoms in superfluid helium (right): theoretical (line) and experiment [15] (points). The kinks at $T = 2.17$ K are the result of the abrupt disappearance of the superfluid component at this temperature.

4. Phonon-roton wing of ZPL

In Ref. [14] the phonon-roton wing of ZPL was measured. This wing is weak as compared to ZPL, and corresponds to single phonon transitions. The spectrum of this transition equals

$$I_{1Ph}(\omega) = \upsilon_\omega (n_\omega + 1). \tag{8}$$

To find this spectrum, one needs to know the DOS $\rho(\omega)$. If we neglect the change in the local dynamic of phonons by the impurity atom and consider the wave numbers k as quantum numbers, then the DOS of the phonon-roton excitations can be represented as

$$\rho(\omega) = (3/k_m^3) \int_0^{k_m} dk k^2 \delta(\omega - \omega(k)),$$

where $\omega(k)$ is the dispersion of phonon-roton excitations of superfluid He. To take into account that k are not exact quantum number sin a liquid, we can replacing the δ-function with a function with a peak of finite width at $\omega = \omega(k)$. Taking for this function a Gaussian with a width linear with respect to k, we obtain

$$\text{DOS} \equiv \rho(\omega) = \left(3/v k_m^3 \sqrt{\pi}\right) \int_0^{k_m} dk k e^{-(\omega - \omega(k))^2/(vk)^2}, \tag{9}$$

v is the characteristic velocity of the deceleration of density waves. The excitation dispersion is shown in Fig. 3 left. We made the DOS calculations for the deceleration velocities $v = 50$ m/s, $v = 100$ m/s and $v = 250$ m/s. The results of calculations are given in Fig. 3, right.

As it was shown above, in liquid in the long-wave limit, the factor $x_{0\omega}^2$ increases in comparison with solids by a factor $\sim \omega_m^2/\omega^2$. At short distances of the order of the interatomic interval, the atomic structure of the liquid and solid phases is similar. Therefore, for large wave vectors and frequencies, the mentioned increase should be less than in the case of low frequencies. The actual structure of the He atoms surrounded the impurity atom including the bubble structure of the surrounded atoms is important

6

in this limit (for bubbles surrounding an impurity atom in liquid helium, see, e.g. [13]). This circumstance can be
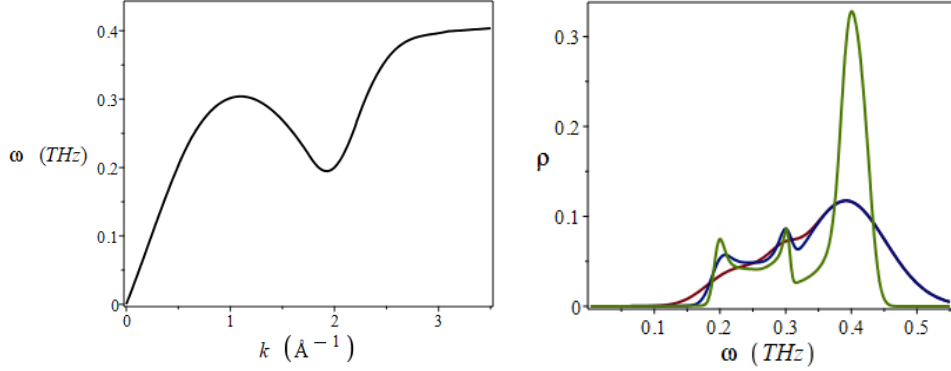

Fig.3. Dispersion (left) of phonon-roton excitations $\omega(k)$ [19] and the DOS ρ of the elementary excitations in the superfluid He (right) for the deceleration velocities $v=50$ m/s (green line), $v=100$ m/s (blue line) and $v=200$ m/s (brown line) calculated in accordance with the equation (9). Minimum(left) and maximum (right) at $\omega=0.19$ THz correspond to rotons, maxima at $\omega=0.3$ THz correspond to maxons, the tail above 0.3 THz in dispersion (left) and the maximum at 0.4 THz in DOS (right) correspond to short-range vibrations of He atoms.

approximately taken into account if we add to the term $x_{0\omega}^2 \propto \omega^{-4}$ an additional small term $\propto \omega_0^{-2}\omega^{-2}$ taking $x_{0\omega}^2 \propto \omega^{-4}\left(1+\omega^2/\omega_0^{-2}\right)$, where the cut-off frequency ω_0 is large as compared to frequencies of roton and phonon. In this approximation

$$I_{1Ph}(\omega)=\alpha\omega^{-1}\left(1+\omega^2/\omega_0^2\right)(n_\omega+1)\rho(\omega) . \tag{10}$$

The spectra of the ZPL and its wing, experimentally observed in [14] at $T=2.1$ and $T=1.35$ are shown in Fig. 4 (left), together with the calculated single-phonon spectra for $v=50$ m/s, $v=100$ m/s and $v=250$ m/s.
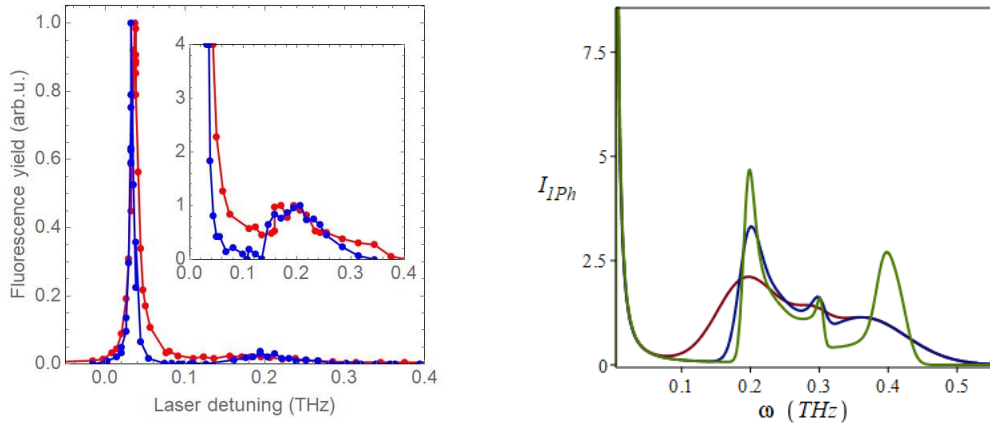

Fig.4 ZPL and its wing in the optical spectrum of Dy atom observed in [14] at $T=2.1$ (red line) and $T=1.35$ (blue line) (left) and calculated in accordance with the equation (10) at $T=2.1$ K (right) single-phonon wing of the ZPL in superfluid He. The velocities $v=50$ m/s (green line), $v=100$ m/s (blue line) and $v=200$ m/s (brown line) are used for calculations. As in Fig. 3, the peak at 0.2 THz corresponds to a roton, the peak at 0.3 THz corresponds to a maxon, and the peak at 0.4 THz is due to a high-energy (large wave number k) tail in the dispersion (Fig. 3, left).

The value of deceleration velocity $v=200$ m/s and the cut-off frequency $\omega_0=1.3$ THz used in calculations of brown curve in Fig. 4 allow us to explain the experimentally observed shape of the

single-phonon spectrum with an almost twofold difference in peak intensities of the roton peak at 0.2 THz and the high frequency phonon peak at 0.35 THz. Relatively higher intensities at $\omega \approx 0.1$ THz in the experiment are due to the multiphonon contributions, which are significant for low frequencies. The contribution made by the additional term ω^2 / ω_0^2 in brackets of equation (10) for all frequencies is less than 0.1. Note that the ratio of the contributions of the roton and the high phonon frequency peaks in the DOS (see Fig. 3, right) is the opposite: the high phonon frequency peak is many time stronger than the roton peak. This large difference in peak intensities in the DOS and in the single-phonon spectrum is mainly due to the amplification factor ω_m^2 / ω^2. This allows us to conclude that the shape of the phonon wing in the spectrum of the Dy atom, observed in [14], directly indicates a strong amplification of the vibronic interaction with low-frequency acoustic phonons in liquids. At the same time, the amplification law works for all phonons with a wave number k less than one or two Å^{-1}. Consequently, the main distinguishing property of the liquid phase - the ability to isochorically change its shape (the Archimedes Principle in the macroscopic limit) is already approximately valid for lengths starting from nm.

5. Summary

The zero phonon line and its phonon-roton wing have been experimentally and theoretically investigated in the optical spectrum of inner shell transition in Dy atom in superfluid helium. It is shown that the temperature dependences of the observed spectrum is a consequence of the redistribution of the superfluid and normal components of the liquid and the temperature dependence of the spectrum of the normal component. It is also shown that the main distinguishing property of the liquid phase is the ability to change its shape isochorically (the Archimedes Principle in the macroscopic limit) - leads to a strong increase in the linear vibronic interaction of impurity atoms with low-frequency phonons and to its divergence as ω^{-1} with a decrease in the frequency ω. As a result, the ZPL of both the superfluid and normal component acquires a finite broadening, and the width of the ZPL of the normal component increases linearly with increasing temperature. The amplification of the vibronic interaction with low-frequency phonons also leads to a strong redistribution of peaks in the phonon-roton wing of the ZPL in favour of the low-frequency ones. Our calculations of temperature changes in the width of the ZPL and the shape of its phonon-rotor wing in the spectrum of the Dy atom are consistent with the experiment.

In conventional liquids, the vibronic interaction is strongly enhanced only for totally-symmetric long-wave acoustic phonons. Therefore, when considering the optical spectrum of Dy atoms in the superfluid ^{4}He, the Jahn-Teller effect was not taken into account. However, our study shows that in liquid crystals, both in nematic (where elongated molecules move freely along the axes of the molecule) and in smectic (where layers can move relative to each other), the vibronic interaction with non-totally symmetrical long-wave acoustic phonons is also enhanced. Therefore, we expect that the Jahn-Teller effect for molecules in liquid crystals is also enhanced compared to the same molecules in ordinary liquids.

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