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Spectroscopy of small helium clusters and ‘nanoscopic’ superfluidity: $\text{He}_N\text{-CO}$, $N = 2 - 20\dots$

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1. Introduction

Studies of clusters attract considerable attention of researchers, because they allow building bridges between the quantum microcosm and the thermodynamic macrocosm. In recent years, much success has been achieved in the study of small clusters consisting of several atoms or molecules bound by van der Waals forces. Such clusters can be comparatively easily produced in the expansion of a supersonic gas jet into a vacuum. It is significant that their spectroscopic research can be conducted in the region of the wide expansion of the jet, where atomic collisions are very rare and the spectral linewidth approaches the Doppler limit. Consequently, the lines of different clusters can be easily resolved, thus permitting tracing the dependence of different physical properties of clusters on their size or composition.

Spectroscopic studies of small helium clusters with an embedded molecule of a chromophore have been developed quite actively in the last few decades. These studies are based on direct measurements of spectra, both purely rotational and vibrational-rotational, in the region of some fundamental vibration of the chromophore, which are then used to determine the effective moment of inertia of the embedded molecule and its vibration frequency shift caused by the interaction with surrounding helium atoms. To date, systems such as $\text{He}_N\text{-X}$, where X is a molecule of OCS [1], CO_2 [2], N_2O [3, 4], CO [5–7], or HC_3N [8] and N smoothly varies from 2 to ≈ 70 , have been studied. These clusters are formed at a finite temperature of the helium jet as low as 0.3–0.1 K. Because the binding energy of all the above molecules—chromophores with helium clusters—is sufficiently small, the resultant clusters are not solid but liquid, and a nonmonotonic dependence of the effective moment of inertia on the number of attached helium atoms is observed for all these molecules. This means that for the number of helium atoms larger than a certain value N , the chromophore rotation with part of the attached helium atoms relative to the remaining helium environment proceeds with sliding. This sliding motion seems to be essentially dissipation-free, because no additional broadening of spectral lines is observed and their widths are such that the molecule appears to accomplish at least 10^5 revolutions without any signs of dissipation. This phenomenon strongly resembles superfluidity and it is this feature that caused a great interest in these investigations. A review of the early experiments was published in [9].

On the other hand, numerous spectral measurements of chromophore molecules in helium droplets [10, 11] (nanodroplets, which contain more than a thousand helium atoms at the temperature ≈ 0.35 K) have been carried out recently. Under these conditions, ^4He is certainly in a superfluid state and the results obtained give the limiting values of the spectroscopic parameters as the cluster size increases. The most spectacular feature that distinguishes superfluid helium from other liquids is the well-resolved rotational structure in

the spectra of molecules embedded into it. As a rule, the measured moment of inertia is several times larger than the moment of inertia in the gas phase. This means that the rotating molecules become covered with a significant helium ‘coat’ during rotation.

An exception is given by light molecules such as HCN and CO, whose moment of inertia increases by only about 20% and the effective size of the ‘coat’ is less than one helium atom. It is significant that in droplets consisting of ^3He with the temperature ≈ 0.15 K, the vibrational lines of the embedded molecules are sufficiently wide and the rotational structure is completely ‘washed out.’ In the experiments with helium droplets consisting of a mixture of ^3He and ^4He , a thermodynamic limit for the appearance of superfluidity was also established, i.e., the minimum number of ^4He atoms ($N_{\min} \approx 60$) necessary for the separation of the isotope solution was evaluated [12].

The properties of small clusters, naturally, depend on the parameters of the interaction between helium and the chromophore molecule, and the nearer their binding energy to the chemical potential of liquid helium (7.5 K), the less the disturbance of the helium environment caused by the molecule. From this standpoint, a unique species is the CO molecule, with its binding energy with helium equal to 9 K; already in the He-CO binary complex, the CO molecule rotates almost freely [13, 14]. At present, the $\text{He}_N\text{-CO}$ system is the only one in which two types of rotation of the embedded molecule are observed: almost free within the cluster; and together with the surrounding helium. The presence of two types of rotation, in addition to being very interesting by itself, allows obtaining additional information on the properties of clusters.

Studying $\text{He}_N\text{-CO}$ clusters is also important for the problem of hydrogen superfluidity. In 1972, Ginzburg and Sobyenin showed theoretically that liquid parahydrogen could pass into a superfluid state at $T \approx 6$ K if it did not become solid at $T \approx 13.6$ K [15]. After this, searches began for metastable states of liquid parahydrogen with sufficiently low solidification points. It is known that the freezing point is strongly reduced in small clusters; indeed, experiments showed that hydrogen clusters with the number of hydrogen molecules $N < 50$ formed in a supersonic gas jet are liquid rather than solid [16]. In clusters of $(\text{paraH}_2)_N\text{-CO}$ with $N = 7 - 17$, a dissipation-free sliding upon the rotation of CO molecules was discovered [17], which indicates a common nature of phenomena observed in these clusters and in small helium systems.

In this report, we consider spectral studies of small $\text{He}_N\text{-CO}$ clusters. Because sufficiently low temperatures are required for obtaining such clusters, only the lowest rotational level of the molecule with an angular momentum $J = 0$ proves to be populated and only transitions from this level, i.e., $J = 1 - 0$, are observed. The frequency of one of these transitions lies near the free rotation frequency of CO (115 GHz); the other transition in small clusters lies in the range of centimeter wavelengths. Two analogous transitions are observed near the fundamental vibration of the CO molecule (in the region of 2100 cm^{-1}). Specifically, it was these transitions that were first investigated in these clusters [5, 6]. Unfortunately, it is impossible to independently determine two rotational constants and the fundamental vibration frequency of CO in the cluster from the positions of these two measured transitions. Therefore, the very interesting results of these works have rather a qualitative

nature, while direct measurements of frequencies of the rotational transitions are necessary for obtaining quantitative characteristics.

2. Experimental methods

As was already mentioned above, helium clusters are formed in a cold supersonic gas jet. To obtain such a jet, a mixture of gaseous helium with a small admixture of chromophore molecules (0.01%) is directed through a supersonic nozzle with a characteristic diameter of the opening of 0.5–1 mm into a vacuum chamber, where the jet expands adiabatically. At a sufficiently large rate of evacuation of the chamber, which ensures the residual gas pressure of the order of 10^{-3} Torr, the efficiency of cooling is mainly determined by the initial pressure and temperature. At the initial pressure 50 atm and the nozzle kept at room temperature, the final temperature of the helium jet reaches 0.3 K, and when cooling the nozzle by liquid-nitrogen vapors, even 0.1 K [5]. To ensure a combination of the necessary fluxes of the gas in the jet with reasonable pumping rates of the vacuum chamber, the nozzle works in the regime of short pulses. For this purpose, a magnetic valve is established before the nozzle entrance; is opened to admit the flow of gas for a period of the order of 1 ms. An oil booster pump with the pumping rate 1000 l s^{-1} creates the necessary vacuum in the chamber at the pulse repetition frequency of the order of 1 Hz.

The spectral studies of helium clusters and binary complexes have been conducted in the infrared (IR) range (McKellar's group, Ottawa), in the centimeter range (Jäger's group, Edmonton), and in the millimeter-wave range (Russian–German group, Troitsk–Cologne) of wavelengths. Because the spectral linewidths in the supersonic jet are close to the Doppler limit, all the measurements have been carried out on spectrometers with coherent radiation sources and high spectral resolution.

A more complicated problem is to reach a sensitivity for registration of weak lines necessary for such experiments. To obtain the necessary temperatures, the content of a chromophore in helium must be low, and the concentration of the formed clusters must be less than 0.1% from that of the chromophore. The sensitivity of the usual single-pass spectrometers is too low for detecting such weak signals; schemes with a multiple pass of radiation through the supersonic jet must be used. In the IR range, a spectrometer based on diode lasers with a toroidal system of mirrors is employed, which ensures more than a hundred passages of the laser beam through the jet [18]. In the millimeter and centimeter ranges, such multipass schemes are unrealistic because of the long wavelength of the radiation, and the only method to achieve the required sensitivity is to pass the jet through a radiation-filled high- Q open resonator. Highly sensitive pulsed Fourier spectrometers in the centimeter range have been developed, which are combined well with open resonators. The installation used in Jäger's group belongs to this type, in particular [19].

Unfortunately, these spectrometers are ineffective in the millimeter-wave range because sufficiently powerful radiation sources with a large frequency tuning are absent at present. In our group, we have developed an intracavity spectrometer [20] based on a domestic wide-range oscillator in the millimeter range, the so-called orotron [21]. The spectrometer developed has its own radiation-frequency-driving open resonator, through which the gas to be

investigated can be passed. As a result, about hundred effective passages of radiation through the jet can be obtained, which ensures a sufficiently high spectrometer sensitivity.

Owing to the high spectral resolution, the spectral lines that refer to different clusters do not overlap and can be analyzed separately. Furthermore, it is very important that the optimum temperature of cluster formation monotonically decrease with increasing the cluster size. Therefore, the intensities of spectral lines pass through a maximum with decreasing the temperature (with increasing the initial pressure of the gas and/or cooling of the nozzle). Based on the position of this maximum, a given spectral line can be referred to a cluster of specific composition.

Of great service in the identification of complex spectra is the double-resonance method. If one of the two transitions in a three-level system is saturated, the populations of all levels change and, correspondingly, the intensity of radiation absorbed in another transition also changes. Consequently, by tuning the spectrometer to the second transition, it is possible to detect the absorption of radiation from an additional source at the first transition based on a change in the signal of the spectrometer. Along with the selection of the pairs of transitions that relate to three-level systems, the double-resonance method allows effectively extending the relevant spectral range and recording transitions from levels whose thermal population is too low to be observed directly. An orotron-spectrometer-based setup for observing double resonance is described in [22].

3. Experimental results

The frequencies of purely rotational $J = 1-0$ transitions in $^4\text{He}_N\text{-CO}$ clusters are given in Fig. 1. All the high-frequency lines have been measured using the orotron spectrometer, and the low-frequency lines in clusters with $N \leq 7$ were measured

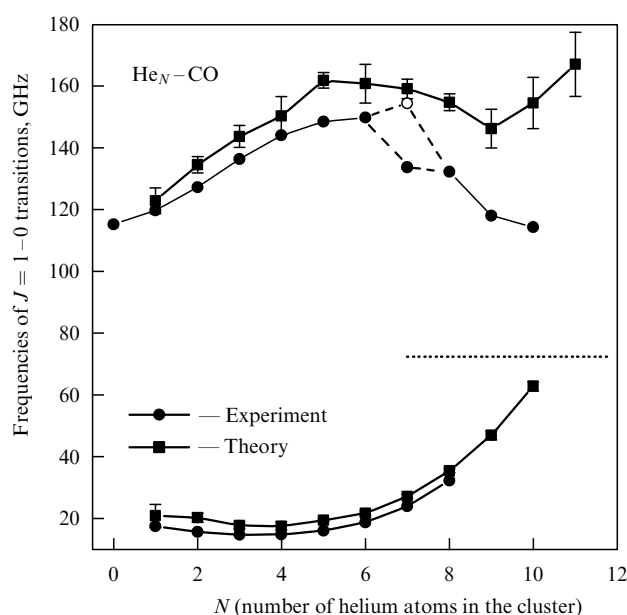


Figure 1. Measured (●) and calculated (■) [23] frequencies of $J = 1-0$ transitions in $\text{He}_N\text{-CO}$ clusters. The frequency of the upper component in the $\text{He}_7\text{-CO}$ cluster (○) was determined from the IR data [5] with allowance for the shift of the CO vibration obtained in [7]. The dotted line shows the value of $2B$ in helium droplets [24].

using the pulsed Fourier spectrometer. The corresponding line in ${}^4\text{He}_8\text{--CO}$ that lies higher than the Fourier spectrometer range is registered on the orotron spectrometer using the double-resonance technique. The low-frequency transitions correspond to the rotation of a molecule together with a helium coat, and their frequency is inversely proportional to the moment of inertia of CO with the attached helium coat. The moment of inertia increases upon attaching up to three helium atoms to the molecule, and then begins decreasing; hence, the sliding upon the rotation of the molecule begins in a ${}^4\text{He}_N\text{--CO}$ cluster that contains four helium atoms. Subsequently, the transition frequency increases smoothly and at $N \geq 6$ exceeds its value in ${}^4\text{He}\text{--CO}$. This means that in clusters with $N \geq 6$, the effective size of the coat is less than one helium atom.

The frequency of high-frequency transitions first increases with increasing the cluster size, passes through a maximum at $N = 6, 7$, and then decreases. In a cluster with $N = 7$, the transition line in the IR spectrum splits into two components. We could register only one line, but its proximity to the position of the lower component predicted on the basis of IR data suggests that the splitting also exists in the fundamental vibrational state of ${}^4\text{He}_7\text{--CO}$.

For describing the observed rotations of a CO molecule in helium clusters, the following qualitative model can be suggested. Because the interaction potential is independent of the azimuthal angle, a helium cluster has an almost cylindrical symmetry. The rotation axis of CO is parallel to the cylinder axis during low-frequency rotations and is perpendicular to it in high-frequency rotations. Because the helium atoms are attracted predominantly to the oxygen end of the molecule [13], the helium environment is deformed near this end; this deformation accompanies the low-frequency rotation of CO and makes a contribution to the measured moment of inertia. As the number of helium atoms increases, the cluster becomes more rigid and its deformation decreases. The transition frequency increases until the first coordination shell is filled up, which is indeed observed in the IR range [5].

In a high-frequency rotation, the role of the helium environment reduces to increasing the anisotropy of the potential of interaction with CO, which leads to an increasing transition frequency. This phenomenon can be described in the framework of the perturbation theory developed for ${}^4\text{He}\text{--CO}$ in [13] using realistic values of the relevant constants. However, in clusters with a filled coordination shell, the difference between the two rotations disappears and their frequencies must be close. Therefore, as the number of helium atoms in a cluster approaches $N \approx 13, 14$, the frequency of the high-frequency transition is reduced.

Figure 1 also gives the results of the calculations of excitations in the ${}^4\text{He}_N\text{--CO}$ system obtained by the diffusion Monte Carlo method [23]. The results of calculations satisfactorily describe the behavior of the low-frequency branch, although the calculated values are somewhat overstated. Things are worse in the case of the high-frequency branch. The experimental frequencies in clusters with $N = 8\text{--}10$ decrease monotonically, whereas the theory predicts their increase in this region. The fundamental question here is whether the transition frequencies in large clusters remain sufficiently high, as the calculations predict, or both rotation branches merge. As an experimental indication of the possibility of the second scenario, we note a doubling of the line of the $J = 1\text{--}0$ transition in the ${}^4\text{He}_{15}\text{--CO}$ cluster observed in the IR range [5].

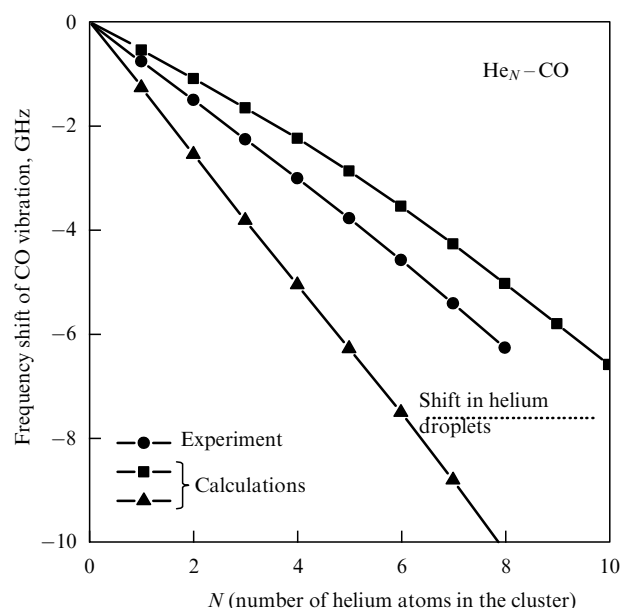


Figure 2. (●) Experimentally determined frequency shift of CO vibration in $\text{He}_N\text{--CO}$ clusters. The results of the theoretical calculations of the shift (■, ▲) are taken from Ref. [23]. The dotted line shows the value of the shift in helium droplets [24].

From a comparison of the microwave and IR spectral data, it is possible to determine the frequency shift of the fundamental vibration of CO in the helium clusters; the data obtained are given in Fig. 2 together with the results of calculations based on different interaction potentials for ${}^4\text{He}\text{--CO}$ [23]. The dependence of the frequency shift on the number of helium atoms in a cluster is almost linear, in contrast to that in helium clusters based on CO_2 , N_2O , and OCS [1–4]. In the latest systems, a change is observed in the sign of the frequency shift at $N = 5$, which is ascribed to the filling of the rigid ring with helium on the equator of these molecules and by the arrangement of the other helium atoms on the periphery of the chromophore molecules. The monotonic character of the shift of the vibration frequency in the clusters based on CO indicates the diffuse nature of the cluster structure, with no essential differences in the arrangement of different atoms. The experimental results are described by the theory qualitatively well, but obtaining a quantitative correspondence requires refining the ${}^4\text{He}\text{--CO}$ interaction potential.

Figure 2 also shows the shift in the frequency of the fundamental vibration of CO in helium droplets that was obtained in [24]. This shift proved to be very small; it approximately corresponds to the value obtained by the extrapolation of our data to the case of ${}^4\text{He}_{10}\text{--CO}$. Because there are no reasons for the change in the nature of the dependence of the frequency shift on the number of helium atoms until the first coordination shell is filled, the shift in these clusters is larger (in absolute value) than in droplets. Therefore, we should either expect a change in the sign of the dependence of the frequency shift on the number of helium atoms in large clusters or suppose that the procedure for determining it used in [24] is not entirely correct.

Figure 3 depicts the dependences of the effective rotational constant B (inverse of the moment of inertia of a molecule with a helium coat) on the number of helium atoms in the cluster for all the ${}^4\text{He}_N\text{--}X$ systems investigated to date, where X is OCS [1], CO_2 [2], N_2O [3], CO , or HC_3N [8]. The

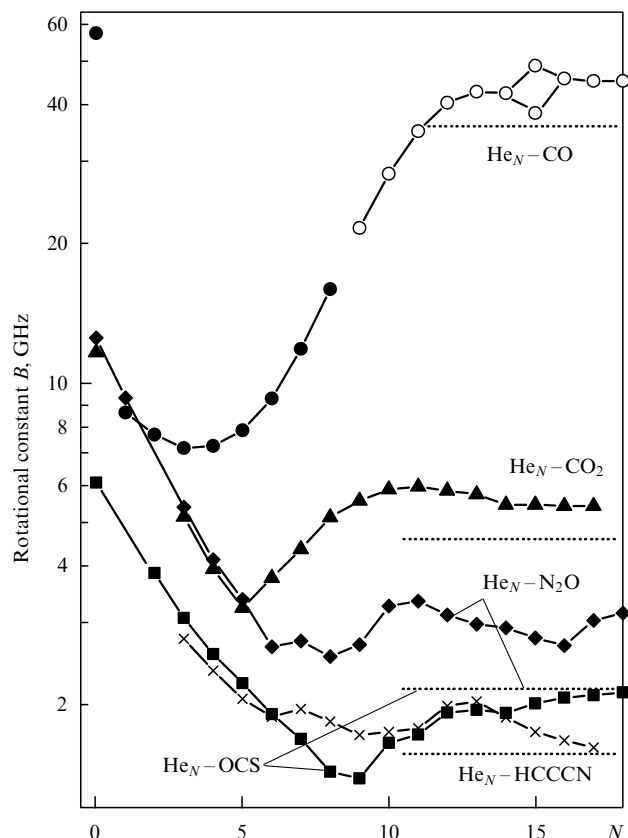


Figure 3. Dependence of the effective rotational constant B of different chromophore molecules on the number N of helium atoms in a cluster. The dotted lines show the limit values of B found in helium droplets.

data for $^4\text{He}_N\text{-CO}$ with $N \leq 8$ have been obtained from the frequencies of the low-frequency rotational transitions [7] and those with $N > 8$, by extrapolation [5] under the assumption of the linearity of the frequency shift of the CO vibration obtained in [7]. In all the systems with the number of helium atoms exceeding a certain value, an increase in the rotational constant (a decrease in the moment of inertia) is observed; this phenomenon appears the earlier, the less the binding energy of helium with the molecule. The ‘record holder’ is the $^4\text{He}_N\text{-CO}$ system, in which the decrease in the moment of inertia begins at $N = 4$, and the effective size of the coat, beginning from $N = 6$, becomes less than one helium atom. At the same time, it can be supposed that other molecules can have a ‘coat’ consisting of several helium atoms and that such a relatively rigid formation rotates inside the remaining helium cluster.

A decrease in the momentum of inertia with increasing the cluster size resembles the Andronikashvili effect [25], consisting in a nonentrainment of superfluid helium with bodies that move in it. It is also significant that with a further increase in the cluster size, the frequency of rotation of chromophore molecules smoothly tends to a limit characteristic of superfluid helium in droplets, which is demonstrated in Fig. 4 using the $\text{He}_N\text{-OCS}$ system with $N = 2\text{--}70$ as an example [1]. A natural question arises regarding the relation of the phenomena observed in small helium clusters to superfluidity. By its properties, helium in a cluster resembles a quantum liquid in which single-particle excitations that can lead to relaxation are absent. Otherwise, an additional broadening of spectral lines would be observed. At the same time, around ten atoms is certainly insufficient for the formation of a quasi-contin-

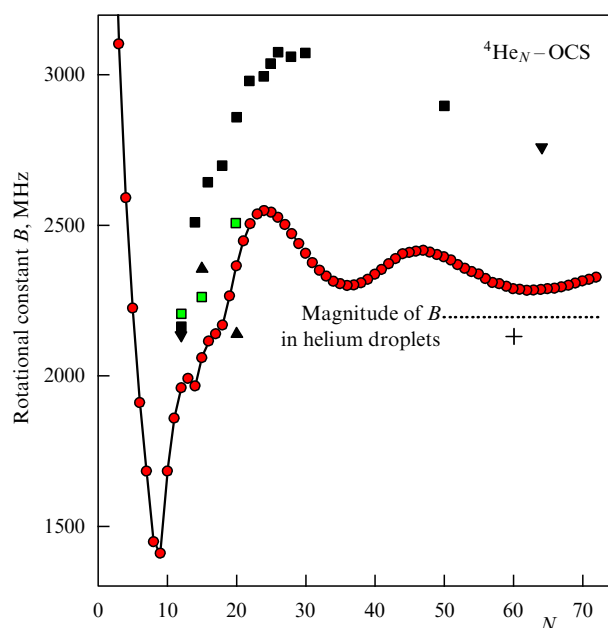


Figure 4. Dependence of the rotational constant B of an OCS molecule on N for $^4\text{He}_N$ clusters [1].

uous spectrum of the Bose type, and the absence of relaxation is most probably related to the discrete character of the spectrum of excitations of a small cluster. Therefore, the phenomena observed in small clusters can be treated in terms of ‘nanoscopic superfluidity,’ which substantially differs from classical superfluidity. Analogous remarks also refer to ‘hydrogen superfluidity,’ which is observed in $(\text{paraH}_2)_N\text{-CO}$ clusters at $N = 7\text{--}17$ [17]. Nevertheless, in a number of works [26, 27], the properties of small helium clusters are treated in terms of fractions of the normal and superfluid helium components, which is unjustified in our opinion.

4. Conclusions

Ya B Zel’dovich once formulated a question: “How many helium atoms are necessary for a droplet to arise?”

It seems that there is no unambiguous answer to this question, and the differences that characterize the physical properties of a droplet begin to manifest themselves at different numbers of atoms. Thus, according to estimations, for a quasicontinuous spectrum of elementary excitations to form, the number of atoms required is of the order of 100, while for the development of sliding upon rotation of a chromophore molecule, only four atoms suffice.

One more question arises: whether analogous dissipation-free sliding upon rotation of a chromophore molecule also exists in fermionic clusters as long as they are so small that excitations of the particle-hole type that can impede the superfluidity are absent. We believe that the answer to this question can be found in the studies of rotational spectra in $^3\text{He}_N\text{-CO}$ clusters. We plan to perform these studies in the near future.

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Single molecules as spectral nanoprobe for the diagnostics of dynamic processes in solid media

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In this report, we briefly review the results of works directed at the development and use of the method of single-molecule spectroscopy (SMS) for investigating dynamic processes in disordered solid-state media.

The first optical experiments on the detection of single-quantum systems were carried out in the mid-1970s in the gas phase with rarefied atomic beams and single ions in electromagnetic traps [1]. An important step in the way of recording spectra of single molecules (SMs) in condensed media was the development of methods of selective laser spectroscopy of impurity centers, i.e., the method of the excitation of fine-structure fluorescence spectra and of burning-out stable spectral holes [2]. We note that both these scientific avenues were developed for the first time at the Institute of Spectroscopy, Russian Academy of Sciences (ISAN).

The first experimental electronic absorption spectra of single chromophore molecules introduced into a solid-state matrix were observed by Moerner and Kador in 1989 [3]. Soon after this, in 1990, Orrit and Bernard [4] showed that

when recording individual spectra of SMs by the method of excitation of fluorescence spectra in the optical range, the signal-to-noise ratio radically increases in comparison with the case of absorption spectra [4]. The possibility of the direct measurement of optical spectra of SMs in solid-state matrices opened the way to setting up various experiments. Since that time, systematic studies of condensed media with the application of the SMS method (see [5] and the references therein) have begun. This area of studies is being vigorously developed now. According to the Web of Science (database of the Institute of Scientific Information, USA), the number of scientific publications devoted to this field in the journals analyzed by the Web of Science was more than 1300 in 2007 alone.

One of the most promising avenues of the SMS method in studies of the properties of condensed media is the use of isolated impurity molecules as spectral nanoprobe for obtaining information on dynamic processes in the local environment of these molecules. The main idea of this approach is that the parameters of an electron transition of an impurity chromophore molecule embedded into a solid-state matrix are very sensitive to the parameters of the local environment of the molecule. The inertialess nature of this spectral probe allows conducting studies with a maximally high time resolution (up to several femtoseconds), and the smallness of the probe dimensions allows obtaining information about the medium on a nanoscopic scale. As a consequence, the optical spectra of impurity molecules contain unique data on the local structure of the matrix and, most importantly, on dynamic processes that occur in the nearest vicinity of such molecules.

The most important advantage of the SMS method is that it allows completely removing the effect of ensemble averaging over a large number of impurity centers, which is inherent in conventional methods of selective laser spectroscopy. Moreover, in contrast to the known methods of investigating the dynamics of solid-state media (based on acoustic or thermodynamic measurements, or on the registration of Raman spectra, inelastic neutron scattering, etc.), the SMS method allows investigating dynamic processes at the level of single molecules and, thus, almost completely removing the averaging over the volume of the sample investigated. This advantage plays a fundamentally important role in the investigations of strongly disordered solid-state media (glass, polymers, polycrystals, nanostructured objects, biological media, and other complex systems), because in such media, as a result of the significant microscopic inhomogeneity of the substance, the parameters and the nature of the local dynamics can differ radically at different points of a sample.

We consider the basic ideas of the low-temperature dynamics of disordered solid-state media in more detail. Numerous studies have demonstrated that at low temperatures, the dynamic characteristics of such media differ significantly from the appropriate properties of crystals. It has also been proved that the observed anomalous properties of disordered media have a universal nature and are practically independent of the concrete structure and chemical composition of the substance. In the specialized literature, diverse disordered media frequently are therefore simply called glasses.

In 1971 [6, 7], a fruitful idea was proposed on the existence of specific elementary low-energy excitations in such disordered solid-state media, namely, of so-called tunneling