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Simulations for the time evolution of instantaneous resonant modes: Creation, annihilation and mode exchange

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Abstract

In this paper, we simulate the time evolution of the instantaneous resonant modes (IRMs), which are quasilocalized, in a model of Ga liquid. Through the simulations, we have observed the whole process from creation to annihilation of each individual IRM, and its lifetime is, therefore, estimated. We also find the exchange in character between an IRM and a non-resonant instantaneous normal mode (INM). The exchange occurs as the two INMs have very close frequencies and share a common central particle.

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1. Introduction

Resonant modes, the low-frequency quasilocalized vibrational excitations, play an important role on the low-temperature properties of glassy materials [1]; however, the microscopic features of the resonant modes are not fully understood [2]. Especially, how the resonant modes are created and annihilated and how they evolve in time are still open questions. Computer simulation is available for studying these problems; however, it takes a quite long computing time for the glassy systems.

Down to the single-particle regime, the short-time liquid dynamics can be exactly described by the instantaneous-normal-mode (INM) approach [3,4], in which the potential energy surface near each configuration is approximated by a harmonic one with the same curvatures. Becuause the anharmonicity of the extraordinarily rugged potential energy landscape of a liquid, the INMs lose their identities via either adiabatically evolving to new INMs of configuration at the next instant or temporarily destructed by mixing two INMs as their frequencies get very close [5]. Thus, each individual INM actually survives for a short lifetime, but with some created and some destroyed during time evolution, the overall INM density of state (DOS) of a liquid, after averaged over configurations, keeps stable.

Encouraged by the recent investigations in terms of inelastic X-ray scattering techniques, the study of microscopic dynamics in liquid metals is continuously an interesting topic [6]. Recently, in a model of Ga liquid, the low-frequency quasilocalized INMs, termed as instantaneous resonant modes (IRMs), have been numerically evidenced [7], and the local structures around the quasilocalization center of an IRM have been reported [8]. The occurrence of the IRMs in this model is attributed to two factors: (A) the well-known curvature dip in the repulsive core of a Ga atom, with the atoms in this shell region being weakly connected in vibration with the central one, and (B) the local volume expansion, which pushes more neighbors of a Ga atom into this shell region. Due to the coorperation of the two factors, the center of a quasilocalized vibration is instantaneously created at a Ga atom, which is more weakly coupled with its neighbors

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than the other atoms. The energy profile along the eigenvector of an IRM can be generally described by the soft-potential model, which is typical for the resonant modes found in a glassy system [9,10]. Thus, the IRMs are quite similar in feature as the resonant modes in glassy systems, except for short lifetimes. In this paper, we will study the time evolution of the IRMs in the Ga-liquid model through tracing the INMs with computer simulation.

2. The simulation

We have performed MD simulations of 500 Ga particles in equilibrium liquid states at pressure about 1 bar; the details of the Ga pseudopotential and our simulations are referred to Ref. [11]. Our model has an accurate fit for the experimental structure factors of Ga liquid. The velocity autocorrelation function $C_v(t)$ at $T = 973$ K obtained from our MD simulations and its power spectrum are presented in Fig. 1. The INM and IRM DOS of our model have been given in Ref. [7]. Shown in Fig. 1 is also the comparison of $C_v(t)$ calculated by the INM DOS with the simulated result [4]. Within 80 fs indicated by the good agreement between the simulated $C_v(t)$ and the INM calculation, the microscopic dynamics of this liquid is exactly described by the INMs. However, because of the imaginary INMs, the agreement breaks down beyond this time interval.

Our simulations for tracing the IRMs include two stages: In the first stage, for finding an IRM, the simulation time step t_0 was set to be 5.45 fs and both particle positions and velocities were collected once an IRM was found. In the second stage, with the collected data as the initial conditions, we traced the INMs both forward and back in time (by reversing the signs of all initial velocities for the later case). During this tracing simulation, the time step is smaller in two orders than the one used in the first stage.

Some variables are required to monitor the INMs during the tracing simulation. Usually, the INM frequencies are used to identify the INMs between two successive instants in a time step, as long as the change of an INM frequency is much smaller than the differences of this frequency from its adjacent ones [5]. However, as two INM frequencies are almost degenerate at an instant, other variables are needed to distinguish them. The chosen variables rely on the characters of the traced INMs. For instance, in a study for the lifetimes of the high-frequency INMs, which are localized only on two particles, the projection of an INM on a particle, defined as $P_{\alpha,j} = |\mathbf{e}_{\alpha,j}|^2$ with $\mathbf{e}_{\alpha,j}$ being the component of the normalized INM eigenvector for this particle, has been used as the tracing variable [5]. According to the characteristics of quasilocalization, an IRM can be identified by the reduced participation ratio s_α of the INM in less than half [7]. Thus, we use both s_α and the largest $P_{\alpha,j}$ of an INM to monitor the time evolution of the IRMs.

3. Results

The time evolution of several INMs in a low-frequency window in the real branch is presented in Fig. 2, in which only the numerical data are shown and there are no continuous lines connecting any two data points at succesive time steps. Because of the small time step, the trajectories of these INM frequencies are generally catched, and the modulation of a trajectory in this low-frequency region varies gently. However, due to the discreteness of the data points, it is hard to tell whether two trajectories cross through each other at some instant during a time step, or are really avoided as in an adiabatic process.

The whole process for the creation and annihilation of an IRM in the model liquid at $T = 973$ K, contrasted with the evolution of a non-resonant, extended INM with a very close frequency, is shown in Fig. 3. Including the trajecto-

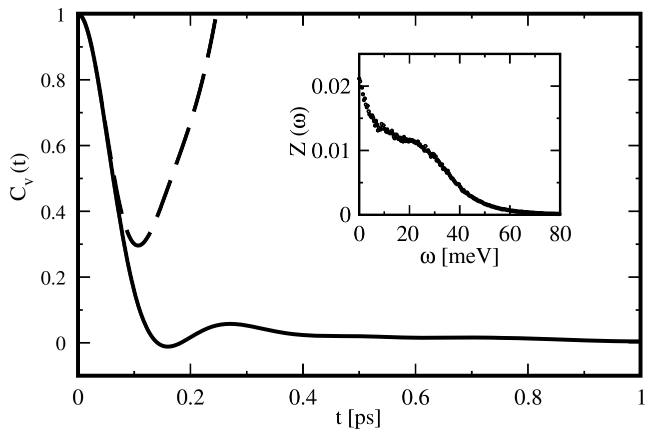

Fig. 1. Velocity autocorrelation function $C_v(t)$ of Ga liquid at $T = 973$ K. The solid line is the result of MD simulation, and the dashed line is calculated with the INM DOS. The inset shows the power spectrum of $C_v(t)$.

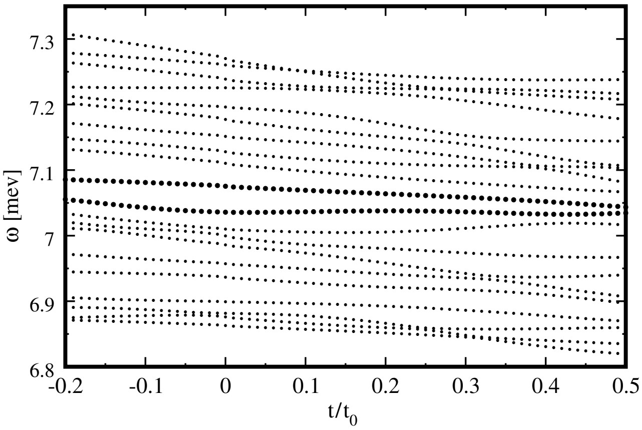

Fig. 2. Trajectories of the INM frequencies in a window of the real spectrum of Ga liquid at $T = 973$ K. The time unit t_0 is 5.45 fs and $t = 0$ indicates the time as an IRM was found in the first stage of our simulaiton. The two INMs explored further in Fig. 3 are indicated by enlarged symbols.

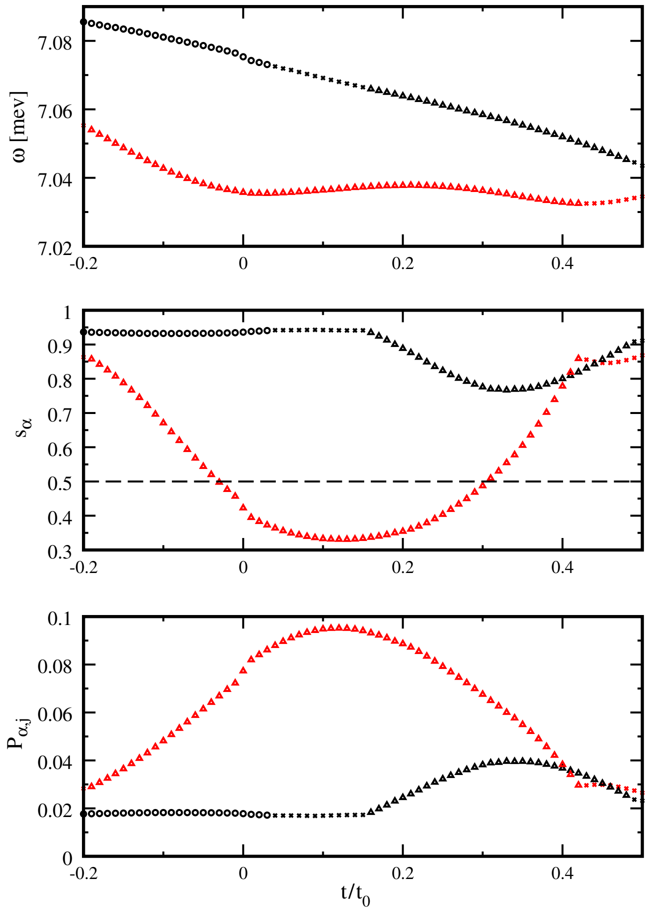

Fig. 3. The creation and annihilation of an IRM (red) in Ga liquid at $T = 973$ K, constrasted with a non-resonant, extended INM (black). The three panels from top to bottom show the trajectories for the INM frequencies, the reduced participation ratios and the eigenvector projections on the central particles of the two INMs. The data are indicated by the symbols of the central particle of the INM at each instant. The circle and triangle are assigned for two specific particles and the cross for the rest particles. (For interpretation of the references in colour in this figure legend, the reader is referred to the web version of this article.)

ries of the INM frequency, s_α and the largest $P_{\alpha,j}$, the data in Fig. 3 are indicated by the assigned symbols for the central particles of the monitored INMs. By the variation of the symbols, how often the central particle of an INM changes with time is manifest. It is clear that at $t = -0.2t_0$ both of the two INMs are extended in space. As time evolves, one INM starts to become quasilocalized, indicated by the descending of s_α from 0.85 down below half and the increasing of $P_{\alpha,j}$ at the same time. Near $t = 0.1t_0$, the value of s_α reaches a minimum and the value of $P_{\alpha,j}$ reaches a maximum. As time evolves further, this IRM is annihilated and recovers back to an extended INM, evidenced by the ascending of s_α and the decreasing of $P_{\alpha,j}$, almost back to their original values. As the IRM is being annihilated, the non-resonant INM, sharing a common central particle with the IRM, tends to be quasilocalized, but this process is uncompleted. Through the time evolution, the lifetime of this traced IRM is estimated to be 1.8 fs.

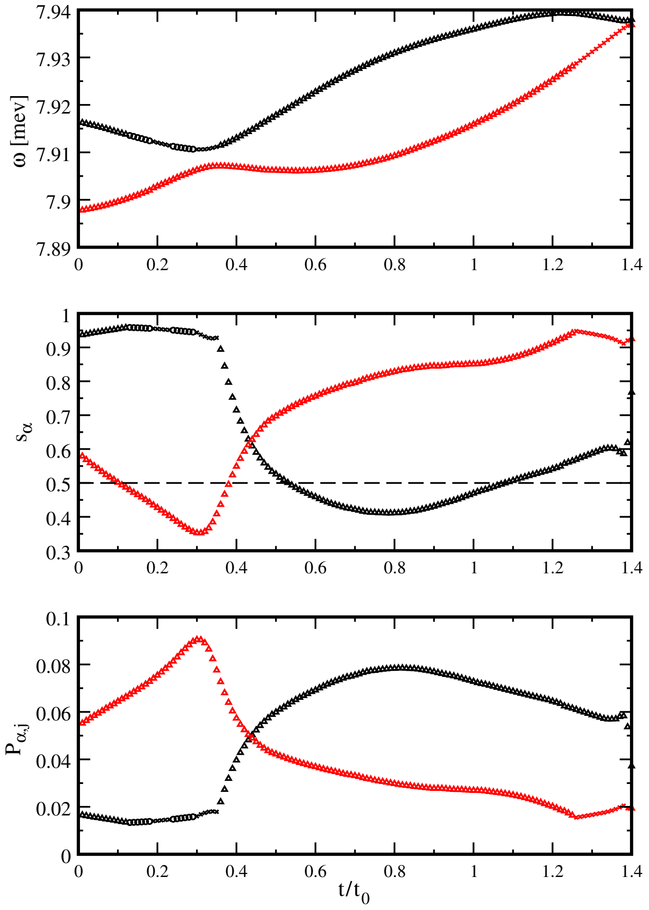

Fig. 4. The character exchange between two INMs with close frequencies in Ga liquid at $T = 323$ K. The meaning of the symbols is the same as that in Fig. 3.

We have observed an exchange in character between an IRM and a non-resonant INM; the process, which occurs in this model liquid at $T = 323$ K, is shown in Fig. 4. As t is less than $0.3t_0$, one of the two INMs is quasilocalized and the other is extended in space. Near $0.3t_0$, the frequencies of the two INMs get very close, but not across each other, and the non-resonant INM changes its central particle to that of the IRM. Once they share the same central particle, the quasilocalization of the original IRM starts to decay; meanwhile a new quasilocalization starts to build up in the other INM. This exchange process continues during an estimated period of 2–3 fs. After this period, the characters of the two INMs are completely exchanged.

4. Conclusion

In this paper, the time evolution of the IRMs, from creation to annihilation, in a model of Ga liquid has been simulated. Through the simulations, the lifetimes of the IRMs are estimated to be of fs order. We also present the numerical evidence for the exchange in character between a IRM and an INM extended in space; this occurs as the two INMs are close in frequency and share a common central particle. This mode exchange is considered as a mechanism to elongate the IRM lifetime.

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References

[1] W.A. Philips (Ed.), Amorphous Solids. Low Temperature Properties, Springer-Verlag, Berlin, 1981.
[2] H.R. Schober, G. Ruocco, Philo. Mag. 84 (2004) 1361.
[3] R.M. Stratt, Acc. Chem. Res. 28 (1995) 201.
[4] T. Keyes, J. Phys. Chem. A 101 (1997) 2921.
[5] E.F. David, R.M. Stratt, J. Chem. Phys. 109 (1998) 1375.
[6] T. Scopigno, G. Ruocco, F. Sette, Rev. Mod. Phys. 77 (2005) 881, and references therein.
[7] T.M. Wu, S.F. Tsay, S.L. Chang, W.J. Ma, Phys. Rev. B 64 (2001) 064204.
[8] T.M. Wu, W.J. Ma, S.L. Chang, S.F. Tsay, Physica B 316–317 (2002) 606.
[9] V.G. Karpov, M.I. Klinger, F.N. Ignat'ev, Sov. Phys. JETP 57 (1983) 439.
[10] U. Buchenau, Yu. M. Galperin, V.L. Gurevich, D.A. Parshin, M.A. Ramos, H.R. Schober, Phys. Rev. B 46 (1992) 2798.
[11] S.F. Tsay, S. Wang, Phys. Rev. B 50 (1994) 108.