Shishkov *et al.* Reply: In the method for enhancing Raman signals that we propose [1], coherent IR laser light, which illuminates a Raman-active molecule, drives oscillations of the molecular electronic subsystem. The driven oscillations parametrically excite coherent nuclear oscillations via the Fröhlich interaction. These oscillations, in turn, modulate the driven optical oscillations of the dipole moment of the electronic subsystem that coherently radiates. This method is based on the theory of Raman phenomena that we develop [1] (see also [2]). The theory obviates the need to introduce uncontrolled phenomenological models such as a nonlinear Hamiltonian or virtual levels. In contrast to the theory used for coherent anti-Stokes Raman scattering (CARS) [3] and Mid-IR assisted CARS [4], we use the time-dependent perturbation theory to solve Heisenberg equations for operators of the electronic dipole moment and the amplitude of nuclear vibrations. In each order of the perturbation theory, all equations are linear and can be solved analytically. This approach principally differs from that used in [3,4], where the perturbation theory is employed to calculate probabilities of transitions between virtual states of the electron subsystem. Our approach also gives conditions for the resonance excitation of nuclei vibrations.

Usually, incoherent radiation is related to the spontaneous Raman effect arising from quantum transitions between virtual levels, while the coherent part of radiation is described classically by $\chi^{(3)}$ as the third order nonlinearity. Our theory allows for distinguishing between *coherent* photons emitted due to the driven motion from *incoherent* photons arising due to the interaction with a reservoir. For this purpose, we do not have to assume that “all molecules are in the same state” as in [5].

In [1], we emphasize a genetic connection between our method and CARS. However, the two methods are distinct. In CARS, the parametric interaction of the electromagnetic wave and molecular oscillations allows for enhancing these oscillations at the frequency difference between *two* incident waves with frequencies ω_{vis1} and ω_{vis2}. In our method, the resonance excitation of the molecular oscillations can be achieved by using a *single* wave with frequency ω_{IR} which is half the frequency of the oscillations. Therefore, the statement of [6] with reference to [3] that our work is not original is surprising—the latter paper does not even mention the phenomenon that we consider.

Unlike in Mid-IR assisted CARS [4], which deals with *IR* active molecules, our method deals with *Raman* active molecules and does not require an intermediate IR active electronic state in the middle between two Raman active electronic states. These two methods deal with different objects and physical phenomena.

In CARS and our method, the difference in estimates is proportional to the ratio $(\omega_0-\omega_{\mathrm{vis1}})(\omega_0-\omega_{\mathrm{vis2}})/(\omega_0^2-\omega_{\mathrm{IR}}^2)$, where ω_0 is the transition frequency between unperturbed electronic states. For nitrogen, $\omega_0 = 12.6$ eV, then for $\omega_{\mathrm{IR}} \sim 0.2$ eV and $\omega_0 \gg \omega_{\mathrm{vis}}, \omega_{\mathrm{vis1}}, \omega_{\mathrm{vis2}}, \omega_{\mathrm{IR}}$, we obtain $(\omega_0-\omega_{\mathrm{vis1}})(\omega_0-\omega_{\mathrm{vis2}}) \sim 0.7(\omega_0^2-\omega_{\mathrm{IR}}^2)$. To evaluate the ratio of coherent and incoherent signals, we use the data of CARS experiments [7,8], where regions with the coherence length up to 1 cm and the waist of the beam w of about 200 μm is used. The corresponding solid angle in which the coherent signal is collected is $\Delta\Omega \simeq \lambda^2/w^2 \sim 2.5\times10^{-5}$. This angle is smaller than the one used in [6] by 2 orders of magnitude. Our estimates are realistic for the settings described.

In [1], we have made errors in numerical values of two parameters of a single nitrogen molecule: for the nuclear vibrational frequency and the electronic transition we have used $\omega_v \approx 100$ meV and $\omega_0 \simeq 3$ eV, respectively, instead of $\omega_v \approx 288$ meV and $\omega_0 \simeq 12.6$ eV, while the Rabi constant $\Omega_{\mathrm{IR}} \approx 10$ meV. The latter, however, can be increased up to $\Omega_{\mathrm{IR}} \approx 50$ meV if one uses a high-power IR laser and focuses it at a spot $\simeq$10 μm. Using these parameters, we obtain $n_b^{\mathrm{coh}} \approx 1.5\times10^{-5}$ and $n_b^{\mathrm{coh}}/n_b^{\mathrm{incoh}} \simeq 25$ compared to $\sim 10^{-3}$ given in [6]. Note that for the application of the Raman response, not only is the enhancement important, but also the practicality of the method. In several situations, using an IR laser can be preferable to using an optical laser. In biological molecules and live tissue, for example, the IR electromagnetic field is less destructive than that in the visible.

Last, the authors of [6] state that the coherent CARS signal depends on temperature. By increasing the density matrix dimension, our method can be readily generalized to include vibrational-rotational transitions and can be used for measuring temperature.

To summarize, we develop a microscopic theory that explains the Raman effect without using phenomenological assumptions. On the basis of this theory, we suggest a new method that is different from that suggested in [6]. This method works in a different frequency range than CARS, and is advantageous compared to CARS in several cases.

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