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Surface Photon Echoes in the Infrared Range

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Abstract. We discuss the possibility of observing photon echoes in the infrared range from a surface monolayer. Three different detection schemes are considered: direct infrared detection, external up-conversion, and in-situ up-conversion. The external up-conversion scheme appears most promising, while the in-situ up-conversion scheme has the advantage of being highly surface specific. Dephasing relaxations of surface vibrational excitations should be measurable.

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Relaxations of various surface excitations are subjects of great importance in surface science. The decay rate and pathway and the coherent phase relaxation of an excitation often play crucial roles in a surface reaction or dynamic process [1, 2]. Dynamics of surface vibrational excitations is of particular interest to many researchers and has attracted considerable research effort in the past two decades [3–14]. In most cases, the experiments strive to attain well-resolved surface vibrational spectra and the width of a vibrational line is assumed to be directly related to the relaxation of that vibration. Theoretical effort to understand such surface relaxation is then focused on relaxation models that can reproduce the measured spectral lineshape [5, 7, 13, 14]. However, it is well known that spectral lines of condensed matter are often dominated by inhomogeneous broadening. In that case, the spectral lineshape contains little information about the relaxation of the excitation. Obviously, transient spectroscopic techniques capable of direct time-resolved measurements of decay of the excitation are needed.

Recently, using the pump-and-probe technique with picosecond infrared laser pulses, Heilwiel and coworkers have made time-resolved studies of the longitudinal (population) relaxations of vibrations for a number of molecular adsorbates on glass [12]. In these experiments, an infrared pump pulse is used to populate a selected vibrational state of the adsorbates and subsequently, a time-delayed infrared probe pulse is used to probe the population relaxation towards thermal equilibrium. Porous samples with high surface-to-volume ratio are often required in order to have an easily detectable signal. Unfortunately, such samples have rather ill-defined surfaces, making the results more difficult to interpret. For a more meaningful comparison with theory, a well-characterized crystalline surface is generally preferred [12a]. Furthermore, the above pump-and-probe method does not yield any information about the transverse (dephasing) relaxation of the vibrational excitation.

In this paper, we explore the possibility of using infrared photon-echoes to study surface vibrational relaxations [15–17]. As is well known, the technique has been widely adopted for studies of relaxations of excitations in bulk samples. Applications of the same technique to surface excitations, however, have not yet been demonstrated. The main difficulty lies in the fact that the total number of excited molecules in a surface monolayer is much smaller than in a bulk. This, together with the lower oscillator strengths of vibrational excitations compared to electronic excitations, results in rather weak IR photon echoes which are difficult to detect because of the lack of sensitive IR detectors. Nevertheless, our estimates show that photon echoes from surface vibrational excitations should be detectable. We consider in this paper three different schemes that could work with a sufficient signal-to-noise ratio: one with a sensitive IR detector,

one with an in-situ up-conversion scheme, and one with an external up-conversion of the signal. In the following section, we first give a brief review on the theory of photon echoes and then extend it to the surface case.

1. Theoretical Background

The theory of photon echoes from a bulk is well known. We extend it here to surfaces. Consider first a monolayer of molecules situated in a refractive-index-matched medium with dielectric constant ε. Let the monolayer be resonantly excited by two short optical pulses, $\mathbf{E}_1(t)$ and $\mathbf{E}_2(t)$, separated by Δt in time.

$$\begin{aligned}\mathbf{E}_1(t)&=\hat{\mathbf{e}}_1\mathscr{E}_1\tau_p\delta(t-t_1)\exp(\mathrm{i}\mathbf{k}_1\cdot\mathbf{r}-\mathrm{i}\omega t)\\ \mathbf{E}_2(t)&=\hat{\mathbf{e}}_2\mathscr{E}_2\tau_p\delta(t-t_1-\Delta t)\exp(\mathrm{i}\mathbf{k}_2\cdot\mathbf{r}-\mathrm{i}\omega t),\end{aligned}\tag{1}$$

where we have approximated the pulses with pulse-width τ_p by δ-functions. The strengths of the pulses are usually expressed in terms of the pulse areas

$$\begin{aligned}\theta_1&=2\gamma_1\mathscr{E}_1\tau_p/\hbar,\\ \theta_2&=2\gamma_2\mathscr{E}_2\tau_p/\hbar\end{aligned}\tag{2}$$

with $\gamma_i=\hat{\mathbf{e}}_i\cdot\boldsymbol{\mu}$, $\boldsymbol{\mu}$ being the electric-dipole matrix element of the resonant transition. Following the work of Hartmann and coworkers, we find the appearance of an induced surface polarization $\mathbf{P}_s(t)$ in the molecular monolayer at $t\sim t_1+2\Delta t$ [15],

$$\begin{aligned}\mathbf{P}_s(t)=&(N_s\boldsymbol{\mu}/2\mathrm{i})\sin\theta_1\sin^2(\theta_2/2)\\ &\times\mathrm{e}^{-2\Delta t/T_2}\tau_p\delta(t-t_1-2\Delta t)\\ &\times\exp[\mathrm{i}(\mathbf{k}_s\cdot\mathbf{r}-\omega t)],\end{aligned}\tag{3}$$

where $\mathbf{k}_s=2\mathbf{k}_2-\mathbf{k}_1$, N_s is the surface density of molecules, and T_2 is the dephasing time of the transition. The echo field generated by $\mathbf{P}_s(t)$ takes the form

$$\begin{aligned}E_{po}(t)&=\frac{\mathrm{i}2\pi k_s}{\varepsilon k_{sz}}[k_{sz}P_{sx}(t)+k_{sx}P_{sz}(t)],\\ E_{so}(t)&=\frac{\mathrm{i}2\pi k_s^2}{\varepsilon k_{sz}}P_{sy}(t),\end{aligned}\tag{4}$$

for the p and s polarizations, respectively. Here, $\hat{\mathbf{z}}$ is chosen as the surface normal and $\hat{\mathbf{x}}-\hat{\mathbf{z}}$ the plane of incidence.

If the monolayer is now placed at the plane boundary surface between two media a and b with dielectric constants ε_a and ε_b, respectively, and the excitation pulses $\mathbf{E}_1$ and $\mathbf{E}_2$ are incident from the side of medium a, then it can be easily shown that the echo field in the reflected direction is given by

$$\begin{aligned}E_p(t)&=\frac{\mathrm{i}2\pi k_s}{\varepsilon k_{sz}}[k_{sz}L_{xx}P_{sx}(t)+k_{sx}L_{zz}P_{sz}(t)]\\ E_s(t)&=\frac{\mathrm{i}2\pi k_s^2}{\varepsilon k_{sz}}L_{yy}P_{sy}(t),\end{aligned}\tag{5}$$

where the macroscopic local field correction factors L_{ii} have the form [18]

$$\begin{aligned}L_{xx}&=2\varepsilon_a k_{bz}/(\varepsilon_b k_{az}+\varepsilon_a k_{bz}),\\ L_{zz}&=2\varepsilon_a k_{az}\varepsilon_b/\varepsilon(\varepsilon_b k_{az}+\varepsilon_a k_{bz}),\\ L_{yy}&=2k_{az}/(k_{az}+k_{bz}).\end{aligned}\tag{6}$$

In the above expressions, $k_a^2=\omega^2\varepsilon_a/c^2$, $k_b^2=\omega^2\varepsilon_b/c^2$, and the boundary condition $k_{ax}=k_{bx}=k_{sx}$ defines the directions of $\hat{k}_a$ and $\hat{k}_b$ of the reflected and transmitted echo waves in media a and b, respectively. Of course, it is also necessary to have the pump fields in $\mathbf{P}_s$ modified because of the boundary effect; $\mathbf{E}_1$ and $\mathbf{E}_2$ should be replaced by $\mathbf{E}_{L1}=\mathbf{L}\cdot\mathbf{E}_1$ and $\mathbf{E}_{L2}=\mathbf{L}\cdot\mathbf{E}_2$, $\mathbf{L}$ being a diagonal tensor with its elements given in (6) [18]. The echo signal strength in the reflected direction is given by

$$\begin{aligned}S(\Delta T)&=(c/2\pi)\int|\mathbf{E}(t)|^2dtdA\\ &\cong\frac{\omega^4}{2\pi c^3}\left|\frac{\varepsilon_a A}{\varepsilon\tau_p k_{sz}}[k_{sz}L_{xx}\int P_{sx}(t)dt\right.\\ &\quad\left.+k_{sx}L_{zz}\int P_{sz}dt]\right|^2(\lambda^2/A)\tau_p\end{aligned}$$

for the p-polarized output and

$$\cong\frac{\omega^4}{2\pi c^3}\left|\frac{\varepsilon_a A k_s}{\varepsilon\tau_p k_{sz}}L_{yy}\int P_{sy}dt\right|^2(\lambda/A)^2\tau_p\tag{7}$$

for the s-polarized output.

We recognize that in the above equations, the product of $\omega^4/2\pi c^3$ and the absolute square term is the radiation rate of the dipole sheet per unit solid angle, and (λ^2/A) is the solid angle of the echo radiation (λ being the wavelength in medium a). Since $\int\mathbf{P}dt\propto\exp(-2\Delta t/T_2)$, we expect $S\propto\exp(-4\Delta t/T_2)$.

For the case of a three-pulse stimulated photon echo, we find an induced surface polarization

$$\begin{aligned}\mathbf{P}_s=&(N_s\boldsymbol{\mu}/2\mathrm{i})\sin\theta_1\sin\theta_2\sin\theta_3\\ &\mathrm{e}^{-2\Delta t_{21}/T_2}\mathrm{e}^{-\Delta t_{32}/T_1}\tau_p\delta(t-t_e)\\ &\times\mathrm{e}^{\mathrm{i}(\mathbf{k}_s\cdot\mathbf{r}-\omega t)},\end{aligned}\tag{8}$$

where

$$\begin{aligned}\mathbf{k}_s&=\mathbf{k}_3+\mathbf{k}_2-\mathbf{k}_1,\\ t_e&=\mathbf{k}_s\cdot(\mathbf{k}_3t_3+\mathbf{k}_2t_2-\mathbf{k}_1t_1)/\mathbf{k}_s^2;\end{aligned}$$

$\Delta t_{21}=t_2-t_1$, $\Delta t_{32}=t_3-t_2$; t_1, t_2, and t_3 are the times at which the three excitation pulses arrive at the surface, and T_1 is the longitudinal relaxation time. The echo signal strength can be obtained from (7) using $\mathbf{P}_s$ in (8). The direction of the reflected echo radiation is determined by the wavevector matching relation, $\mathbf{k}_{s,\parallel}=\mathbf{k}_{3,\parallel}+\mathbf{k}_{2,\parallel}-\mathbf{k}_{2,\parallel}$, along the boundary surface.

To estimate the signal strength, we assume a pulsewidth $\tau_p=10^{-12}$ s and a dipole moment for molecular vibration $\mu_{ab}=10^{-19}$ esu (0.04 Debye). For $\theta_i\sim 1$ in (1), the infrared pump pulse fluence is about 15 mJ/cm^2, which can be obtained by difference-frequency generation or optical parametric amplification in a nonlinear crystal ($\sim$150 μJ focused to 1 mm^2). Consider now the two-pulse surface photon echo described by (8). If we take the following as the representatives of experimentally achievable parameters,

$$\sin^2(\theta_{p1})=1,$$

$$\sin^4(\theta_{p2}/2)=1,$$

$$A=1\ \text{mm}^2=0.01\ \text{cm}^2,$$

$$N_s=4\times 10^{14}/\text{cm}^2,$$

$$\lambda_{IR}=2\pi c/\omega_{IR}=3\ \mu\text{m}=3\times 10^{-4}\ \text{cm},$$

$$\mu_{ab}=10^{-19}\ \text{esu},$$

$$T=10^{-12}\ \text{s},$$

$$\varepsilon_a=\varepsilon=1,$$

$$\varepsilon_b=10,$$

we find from (8),

$$S(\Delta t=0)\sim 1\times 10^{-10}\ \text{J/pulse}$$
$$\sim 2\times 10^9\ \text{photons/pulse}.$$

Such a signal should be detectable. We discuss in the next section various ways of detecting surface photon echoes.

2. Detection of Infrared Surface Photon Echoes

We discuss here three different methods that could be employed to detect surface photon echoes (or more generally, coherent transient effects) in the infrared: a) direct measurements using sensitive infrared detectors; b) up-conversion of the infrared signal to visible in a nonlinear crystal; c) in-situ up-conversion of the signal with surface infrared-visible sum-frequency generation.

2.1. Direct Measurements with Infrared Detectors

There exist a number of commercially available sensitive detectors such as liquid-nitrogen-cooled HgCdTe detectors or liquid-helium-cooled Si:Sb detectors. They have a noise equivalent power of the order of 10^{-11} to 10^{-16} W (Hz)$^{-1/2}$ and a response time in the range of 10^{-4} s. The minimum detectable pulsed signal is then around 10^{-13}–10^{-18} J/pulse. Thus our estimated photon echo signal of 10^{-10} J/pulse appears to be easily detectable.

A serious drawback of this detection scheme, however, comes with the slow response of the infrared detectors. The 10^{-4} s response time makes gating of a detector to suppress diffusive scattering of the pump beam impossible. About 10^{-6}–10^{-7} of the incoming radiation is diffusely scattered into the detector. For a 100-μJ pump pulse, this would mean that $\sim 10^8$–10^9 noise photons will show up in the detector from diffuse scattering, while the estimated maximum echo signal is also of 10^8–10^9 photons. Thus the detection of the echo signal could be difficult. Obviously, it is more advantageous to employ detection schemes that allow gating in time to discriminate diffuse scattering.

2.2. Detection by Up-Conversion in a Nonlinear Crystal

Infrared-visible sum-frequency generation (SFG) in a nonlinear crystal is an effective up-conversion scheme to detect weak infrared signals [19, 20]. It has been used successfully in many experiments of infrared fluorescence or emission spectroscopy. With a sufficiently intense pump beam, the efficiency of this up-conversion process can be higher than 10%. Since the process requires simultaneous presence of the IR signal and the visible pump pulse in the nonlinear crystal, the pump pulse can provide an effective gating in the detection of the desired IR pulsed signal. Thus the IR-visible up-conversion scheme should be a very promising technique for experimental studies of surface IR photon echoes.

For efficient SFG, collinear phase matching in the nonlinear medium is required, and the length of the medium should be maximized. If the pump and signal pulses are very short, then the medium length is normally limited by the group velocity mismatch of the two pulses. We consider the use of $LiNbO_3$ as the nonlinear crystal. For a pulsewidth $\tau_p=1$ ps, the group velocity mismatch of the two pulses at 0.53 μm and 3.4 μm limits the copropagation length of the pulses in the crystal to 1 mm. Fortunately, the nonlinear susceptibility of $LiNbO_3$ is fairly large ($d_{eff}=2\times 10^{-8}$ esu). To achieve an up-conversion efficiency, η = output SF photons/input signal photons, of

10%, we only need a pump beam intensity of 120 MW/cm^2. This corresponds to a pump pulse energy of 1.2 μJ if the pump beam with $\tau_p = 1$ ps is focused to 1 mm^2 in the crystal. Such a pump pulse is readily obtainable.

Assuming a surface infrared photon echo signal of 10^8 photons per pulse, a 10% up-conversion efficiency, and a 1% detection efficiency of the sum-frequency output, we should then expect to observe 10^5 counts per echo pulse. This means a signal-to-noise ratio better than 10^8 since the noise in a gated detector can be less than 1 count per 10^3 pulses. Such a large signal-to-noise ratio should make the measurements of surface IR photon echoes fairly straightforward. The SFG up-conversion technique certain appears much superior than direct detection by an infrared detector.

2.3. *Detection by In-Situ Up-Conversion*

Neither of the detection schemes discussed above can discriminate emission from a surface against that from the surrounding bulk. Their applications are therefore limited to cases where surface photon echoes can be distinguished from bulk emission and scattering in time and frequency. Otherwise, a surface-specific detection scheme is needed.

Recently, IR-visible sum-frequency generation has been proven to be an effective spectroscopic tool for IR surface spectroscopy [21, 22]. It is highly surface-specific and sensitive to submonolayers of molecules. If we recognize that this surface SFG is merely an up-conversion process to detect the coherent resonant transverse excitation induced by the incoming IR radiation, then obviously the same process can be used to detect the coherent transverse excitation associated with the surface echo formation. Thus, the in-situ up-conversion at the surface can be a surface-specific method for detection of surface photon echoes. Assuming that the areas of θ_i of the exciting pulses are chosen to optimize the echo signal, we expect that the ratio of the resonant transverse excitation associated with echo formation to the direct infrared excitation is

$$[\varrho_{21}(\Delta t)]_{\text{echo}}/[\varrho_{21}]_{\text{IR}} \gtrsim \exp(-2\Delta t/T_2).$$

Since it has been demonstrated that the direct infrared resonant excitation of a surface layer can be detected with a signal-to-noise ratio better than 10^3, the detection of surface photon echoes with this in-situ up-conversion scheme is certainly possible [21, 22].

In comparison with the scheme of external up-conversion in a nonlinear crystal, the present scheme is definitely inferior in the possible signal-to-noise ratio. This is mainly because the effective interaction length for up-conversion is much smaller in the present case (~ 10 Å for a surface monolayer as compared to 1 mm in a nonlinear crystal). The in-situ up-conversion of the echo formation is actually more efficient per unit length than the external up-conversion because the latter requires first the conversion of the surface resonant transverse excitation to IR radiation to be emitted from the surface and then the mixing of the IR radiation with the visible pump beam in the nonlinear crystal. However, the gain in efficiency ($\sim 10^7$) per unit length from in-situ up-conversion is not enough to overcome the loss in efficiency ($\sim 10^{-12}$) due to the much shorter interaction length. As a result, the in-situ up-conversion scheme is expected to have an inferior signal-to-noise ratio ($\sim 10^{-5}$ smaller) than the external one.

3. Conclusion

In summary, vibrational photon echoes from a surface monolayer, and IR surface coherent transient effects in general, should be observable with either a sensitive infrared detector or an up-conversion detection scheme. Bulk scattering of the exciting pulses is expected to be much stronger than the surface echo signal. This suggests that gating in the detection of the signal would be most important. Since we anticipate the dephasing relaxation time to be in the ps and sub-ps regime, the slow response of available infrared detectors makes gating impossible. Up-conversion, on the other hand, is inherently an optical gating process, and therefore is naturally suited for detection of surface photon echoes. The external up-conversion scheme converting the IR echo signal in a nonlinear crystal is most promising because the high conversion efficiency could lead to a very high signal-to-noise ratio. However, it is not capable of providing discrimination against bulk emission. The in-situ up-conversion at the surface has the advantage of being an inherently surface-specific process. Although the signal-to-noise ratio is worse because of the greatly reduced conversion efficiency, this latter scheme will be far superior in cases where bulk emission is overwhelming.

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