THREE-PHOTON INTERACTION IN OPPOSITELY MOVING WAVES AND THE STARK EFFECT IN POTASSIUM VAPOR

V. M. ARUTYUNYAN, N. N. BADALYAN, V. A. IRADYAN, and M. E. MOVSESYAN

Erevan State University; Institute of Physics Research

Submitted May 21, 1970

Zh. Eksp. Teor. Fiz. 60, 62–65 (January, 1971)

The three-photon interaction process in potassium vapor was investigated experimentally in a direction opposite to the propagation of an intense wave. It was found that there was no parametric broadening and there is a considerable enhancement of the three-photon interaction. An intensity-dependent shift is observed for the three-photon scattering line and is connected with the Stark effect in the intense field of the incident wave.

IN earlier communications[1,2] we reported observation of three-photon resonant interaction in rubidium vapor following excitation of chloroprene by the first SRS Stokes component (detuning from resonance 50 cm^{-1}), and in potassium vapor following excitation of chloroform by the second SRS Stokes component (detuning from resonance 21 cm^{-1}). Attempts to observe the three-photon interaction in potassium vapor following excitation by the first SRS Stokes component of nitrobenzene gave no affirmative results[3,4], although a larger effect was to be expected in this case because of the proximity of the resonance (detuning 10 cm^{-1}). In[2] we observed broadening of the spectral lines following passage of intense radiation through a resonant medium (potassium vapor). It was also noted there that such broadening is connected with four-photon parametric interaction. This question was considered theoretically in greater detail in[5], where it was shown that the three-photon interaction is strongly suppressed in the propagation direction of the intense wave, owing to the parametric broadening of the spectrum. In a direction opposite to that of the propagation of the intense wave, there is an appreciable enhancement of the three-photon interaction, there is no parametric broadening, and this should make it possible to determine the line positions more accurately.

The high-frequency Stark effect in a light field on the absorption lines of potassium was observed in[6] and in[7] on the emission lines. In mercury vapor this effect was observed on emission lines under conditions quite far from resonance (detuning 570 cm^{-1})[8].

In the present study we investigated the process of three-photon interaction in a direction opposite to the propagation of the intense wave, as well as the dependence of the Stark shift of the three-photon scattering line on the light-wave intensity.

The experimental setup is shown in Fig. 1. The laser operated in the giant-pulse regime with an emission power ~50 MW. The vapor was excited by the first Stokes component of SRS of nitrobenzene (13053 cm^{-1}) and the second Stokes SRS component of chloroform (13064 cm^{-1}). The power density and the Stokes SRS component amounts to ~20 MW/cm^2 prior to entry into the cell with the potassium, the beam diameter is 1.5 cm, and the divergence angle ~0.5°. The strong SRS component radiation passing through the cell with the potassium was reflected from an ordinary glass plate, again entered the cell, and was then diverted by a plate to the spectrograph slit, with the aid of prisms. For intensity-control purposes, part of the incident radiation was directed to the upper part of the spectrograph slit. Thus, both the "strong" SRS radiation and the "weak" reflected radiation were simultaneously present in the cell. Preliminary experiments have shown that such "weak" radiation does not produce three-photon interaction by itself. The intensity of the incident radiation was varied with a stack of glass plates. The experiment was performed with a potassium cell 30 cm long at a vapor pressure 1.5 mm Hg. The spectra were registered with an ISP-51 spectrograph (dispersion ~30 Å/mm in the region of 7600 Å).

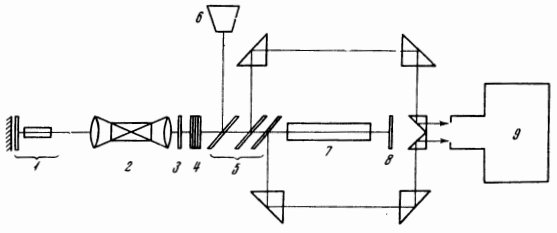

FIG. 1. Diagram of experimental setup: 1—laser, 2—cell with chloroform or nitrobenzene; 3—FS-7 filter, 4—stack of glass plate, 5—beam-splitting plates, 6—calorimeter, 7—cell with potassium vapor, 8—reflecting glass plate, 9—spectrograph.

The spectra registered in accordance with the described scheme have shown that actually there is strong amplification at the frequency of the three-photon interaction when the "weak" and "strong" radiations propagate in the media opposite to each other. Figures 2a and 2b show by way of an example two of the obtained spectra. These spectra show clearly a strong line of three-photon scattering and the absence of broadening of the incident-radiation line. This confirms the assumption that the broadening in the "forward" direction is due to parametric processes that suppress the three-photon interaction in this direction. On most spectra, the intensity of the three-photon line is larger than the intensity of the line of the incident "weak" radiation after passage through the vapor. In "forward" observation, such an intensity ratio was never observed. This indicates that the obtained spectra are not the result of simple "backward"

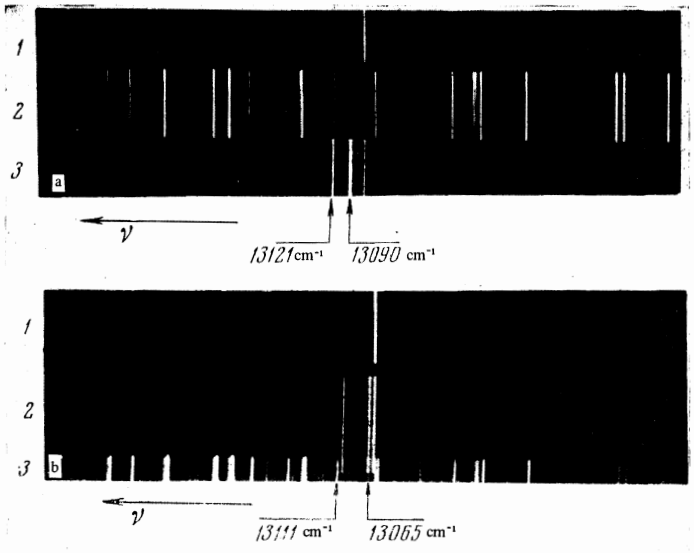

FIG. 2. Excitation of vapor by the second SRS Stokes component of chloroform (a): 1–spectrum of incident radiation, 2–spectrum of iron, 3–spectrum of radiation after the interaction (the frequencies of the electronic Raman scattering and of the three-photon scattering are indicated). Excitation of vapor by the first SRS Stokes component of nitrobenzene (b): 1–spectrum of incident radiation, 2–emission spectrum after interaction with the vapor (the frequencies of the electronic Raman scattering and of the three-photon scattering are indicated), 3–spectrum of iron.

reflection of the picture observed in the propagation direction of the "strong" radiation.

Control experiments have also confirmed the assumption that the three-photon interaction is enhanced in the propagation direction of the "weak" radiation as a result of the interaction of the "strong" and "weak" waves as they move opposite to each other. As a control, the beam was split ahead of the cell, its weak part was directed with the aid of a prism to the cell from the back, in the direction opposite to that of the more intense part of the beam. In this case we observed an amplification of the three-photon interaction line in the spectra registered in the direction of the "weak" beam. When the "weak" beam was blocked in such a system, no three-photon interaction line was observed in such a system in the spectrum of the back-scattered radiation.

The described scheme was used to register the line of three-photon interaction in potassium excited by the first SRS Stokes component of nitrobenzene (Fig. 2b). As indicated, this effect could not be observed in the "forward" direction (even with a cell 1 m long).

The obtained relatively-narrow lines in the spectra made it possible to measure more accurately the frequencies of these lines (within 0.5 cm^{-1}, spectral width of the slit 0.3 cm^{-1}). The results revealed a distinct intensity-dependent shift of the three-photon scattering line, which we attribute to the Stark phenomenon in the intense field of the incident wave. As follows from the theory, the lines shift towards higher frequencies.

The dependence of the three-photon scattering line shift on the intensity of the exciting radiation (second SRS Stokes component of chloroform) is shown in Fig. 3. The magnitude of the shift depends linearly on the

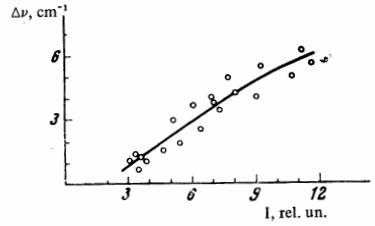

FIG. 3. Shift of three-photon scattering line vs. intensity.

pump intensity. A certain deviation from linearity is observed at larger values of the exciting-radiation power. An accurate knowledge of the effective intensity of the second SRS Stokes component of chloroform within the interaction volume is quite difficult, since it calls for the knowledge of the spatial and temporal distributions in the radiation pulse in the interaction volume. However, one can assess quite accurately the power of the radiation interacting with the potassium vapor, from the value of the Stark shift of the three-photon emission line. The Stark shift of the three-photon interaction line, in accordance with the formula for the quadratic Stark effect, is equal to $(1/2)\epsilon\xi$, where $\xi = 10^{-1} I f_{ik}/\epsilon^2$ [2] (f_{ik} is the transition oscillator strength, I is the flux of number of photons per cm^2 per unit time, and ϵ is the detuning from resonance). A shift of 6 cm^{-1} corresponds to a Stark-shift parameter $\xi = 0.6$. Calculation in accordance with the indicated formula for ξ leads to an intensity $I \sim 10^{26}$ photons/cm^2sec or ~ 30 MW/cm^2.

When potassium vapor is excited by the first SRS Stokes component of nitrobenzene, a shift of the three-photon scattering line is likewise observed, but no direct measurements were made by us in this case.

The spectra of the scattered radiation (on the high-frequency side) contain also an intense line at 13121 cm^{-1} (Fig. 2a) and 13111 cm^{-1} (Fig. 2b), due to stimulated electronic Raman scattering from the $4P_{3/2}$ level to the $4P_{1/2}$ level[2,4]. Within the limits of the accuracy of our measurements, there is no shift of the electronic SRS line. This is apparently connected with the fact that, first, the shift predicted by the theory for the electronic SRS line, at the same intensities, should be half as large, and second, the process of electronic Raman scattering occurs after the three-photon scattering process at lower effective values of the field intensity.

[1] N. N. Badalyan, V. A. Iradyan, and M. E. Movsesyan, ZhETF Pis. Red. 8, 518 (1968) [JETP Lett. 8, 316 (1968)].

[2] V. M. Arutyunyan, N. N. Badalyan, V. A. Iradyan, and M. E. Movsesyan, Zh. Eksp. Teor. Fiz. 58, 37 (1970) [Sov. Phys.-JETP 31, 22 (1970)].

[3] P. P. Sorokin, N. S. Shiren, J. R. Lankard, E. C. Hammond, and T. G. Kazyaka, Appl. Phys. Lett., 10, 44 (1967).

[4] M. E. Movsesyan, N. N. Badalyan, and V. A. Iradyan, ZhETF Pis. Red. 6, 631 (1967) [JETP Lett. 6, 127 (1967)].

[5] V. M. Arutyunyan, E. G. Kanetsyan, and V. O. Chaltykyan, Zh. Eksp. Teor. Fiz. 59, 195 (1970) [Sov. Phys.-JETP 32, 108 (1971)].
[6] A. M. Bonch-Bruevich, N. N. Kostin, V. A. Khodovoĭ, and V. V. Khromov, ibid. 56, 144 (1969) [29, 82 (1969)].
[7] Yu. M. Kirin, D. I. Kovalev, S. G. Rautian, and R. I. Sokolovskiĭ, ZhETF Pis. Red. 9, 7 (1969) [JETP Lett. 9, 3 (1969)].
[8] P. Platz, Appl. Phys. Lett., 16, 70 (1970).

Translated by J. G. Adashko
8