The notation in Eq. (12) is the same as that in Ref. 10. The results of specific numerical calculations carried out using the above scheme will be considered in a separate communication.

Our work has thus provided a systematic analysis of the spectra of the vibrational states ν_1 of three spherical top molecules for which a study has been made of the features of the rotational structure associated with changes in the relative positions of the vibrational states near ν_1. This analysis shows that interpreting the spectra of the ν_1 band and calculating the molecular parameters one has to use a model approach based on a simultaneous consideration of a set of interacting vibrational states.

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Inertialess metal glow produced by picosecond pulses

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A new phenomenon, inertialess glow of a metal produced by picosecond pulses, has been observed and is theoretically explained. The glow is due to the production of hot electrons in the metal. This phenomenon can serve as a basis for the investigation of the relaxation kinetics of the electron and phonon subsystems in metals, and can also be used for inertialess conversion of near and far IR picosecond pulses into visible light. This uncovers prospects for expanding the spectral range of modern photoelectric recorders.

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According to a hypothesis advanced in Ref. 1, temperature-induced glow, due to heating of the electron gas of the metal, should appear under certain conditions in addition to the photoemission and thermionic emission induced by ultrashort pulses in a metal.[2] At sufficient laser pulse-intensity the electron temperature T_e can reach several thousand degrees, and a substantial fraction of the radiation will be in the visible region of the spectrum. The glow follows without inertia the pulse waveform if T_e is not subject to inertia delay. In Ref. 1 were reported preliminary experimental results that confirmed indirectly the advanced hypothesis. The time resolution of the measurements in Ref. 1, however, was worse than 10^{-9} sec at a laser pulse duration $\sim 10^{-11}$ sec, so that no direct confirmation of the hypothesis could be obtained.

We report in this paper experimental results ob-

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tained with the aid of the "Agat" image-converter streak camera at a high time resolution (10^{-12} sec). We analyze and discuss the results, and consider the conditions that must be satisfied by the parameters of the metal and of the laser pulse in order to ensure inertialess glow. We analyze the possibility of using this phenomenon to investigate the relaxation kinetics of the electrons and of the lattice in metals, as well as to study inertialess conversion of picosecond IR laser pulses into visible light.

1. EXPERIMENTAL RESULTS

Experimental investigations of the metal-surface glow were made using the optical system shown in Fig. 1. A neodymium laser with passive mode locking by a solution of dye No. 3955 in nitrobenzene generated a train of 15–20 pulses with average duration ~10^{-11} sec, total duration 100–150 nsec, and average energy per pulse 10^{-4}–10^{-3} J. We note that if the *a* specially chosen active element and a thin-layer flow-through cell and an internal opaque mirror are used in the layer, high temporal stability of the laser is ensured. The laser beam was focused on samples comprising films of various metals (copper, silver, gold) and of varying thicknesses. The irradiated region was projected by condenser *14* and lenses *16* on to the slit of the "Agat" streak camera *17*. The sensitivity of the streak camera was 10^{-10} J during the pulse time. Care was taken in the experiments to prevent stray laser radiation at the fundamental frequency and at higher harmonics from entering the streak camera. This was done with filters *15*. The streak camera was triggered by one of the first pulses of the train.

Figure 2b shows a typical streak photograph of the glow pulse, obtained by exposing a silver film 10^{-4} thick by one of the first pulses of the train. For comparison, Fig. 2a shows streak photographs of a laser pulse (likewise one of the first in the train). The duration of a single laser pulse was 20 psec. Reduction of the streak photographs showed equality, within the limits of the measurement error, of the laser pulse and glow pulse durations. Similar results were obtained also with copper films.

In addition to streak-photograph investigations, we

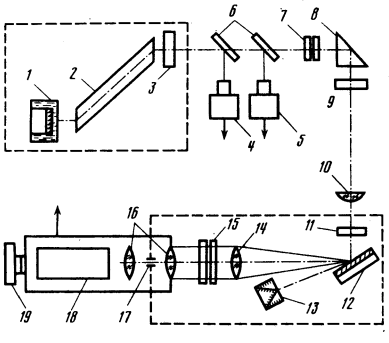

FIG. 1. Optical measurement setup: 1-*Q* switch, 2-active element, 3-exit mirror, 4-FK-2 photocell, 5-calorimeter, 6-beam-splitting plates, 7-attenuators, 8-turning prism, 9, 11-IR filters, 10-lens, 12-sample, 13-Wood's cone, 14-condenser, 15-selective filters, 16-objectives, 17-streak-camera slit, 18-image converter, 19-photographic camera.

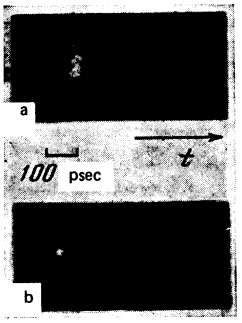

FIG. 2. Streak photographs of laser pulse (a) and of glow of silver (b).

investigated also the glow spectrum, as already indicated in Ref. 1. The glow spectrum was obtained by a photoelectric procedure.[1] A continuous spectrum was observed visually; the color temperature was determined by means of three points, assuming a Planck distribution. For the laser pulse parameters indicated above, the color temperature was 3.5×10^3 K±10% (for silver and copper).

The experiments show thus that at the given laser-pulse and metal-film parameters the observed glow is connected with the electron temperature and follows the laser pulse without delay. However, a more accurate measurement of the shape and duration of the pulse must be carried out under experimental conditions that will be set forth below.

The laser-radiation power density q_d on the film surface had in our experiment a certain stable value q_0 (±15%) in the range 10^9–10^{10} W/cm. With decreasing laser-pulse power density (within one order of magnitude) the glow power dropped below the streak-camera sensitivity threshold. The reason is that the measurements were made with a laser-emission wavelength $\lambda=1.06$ μm and with a camera oxygen cathode having maximum sensitivity in the near infrared. The need to "cut off" the spectrum of the scattered radiation at $\lambda=1.06$ μm greatly decreased the sensitivity of the streak camera and the reception spectrum width. At $q_d>q_0$ the glow spectrum acquired a characteristic fall-off due apparently to violation of the third condition for zero delay [see (5)], inasmuch as measurements have shown that the electron temperature is relatively low and the ion temperature can be appreciably raised on account of the "accumulation effect" under conditions when a pulse "train" is produced. (The gist of the "accumulation effect" is that the characteristic time of film cooling on the glass substrate is commensurate with the time distance between the separate pulses in the train.)

Thus, measurements with a laser of longer wavelength or with a streak-camera cathode having maximum sensitivity in the visible region of the spectrum (antimony-cesium), as well as with a single laser pulse, can extend the dynamic range of the measured laser power density by more than 1–2 orders of magnitude.

The measured pulse time waveforms were reduced by using the Planck distribution. In fact, if we consider a situation wherein the electron temperature follows without delay the waveform of the laser pulse,

then it is obvious that the intensity of the light emitted by these electrons is likewise inertialess. Assume that the electron gas of the metal is in thermal equilibrium with absolute blackbody radiation. Then the radiation energy flux density $I(\omega_i \theta t)$ from the surface of the metal into a unit solid angle, at an angle θ to the normal and in a unit interval of the frequency ω, is given by[3]

$$I(\omega,\theta,t)=(1-R)\frac{\hbar\omega^3}{4\pi^3c^2}\cos\theta\left(\exp\left(\frac{\hbar\omega}{kT_e(t)}\right)-1\right)^{-1}, \quad (1)$$

where R is the coefficient of reflection of light of frequency ω from the metal surface, and c is the speed of light. We note that this conclusion, which is valid under the condition $l_T > \varkappa^{-1}$ (where l_T is the heating depth and $\varkappa^{-1}$ is the absorption length of the visible frequencies) is in fact a general one and does not depend on whether the metal is in equilibrium with the radiation or not.

Expression (1) yields therefore a one-to-one correspondence between the measured laser pulse waveform and the glow pulse shape. It must be noted that these measurements call for sensitometric calibration measurements of the streak-camera screen and of the photographic film.

2. ANALYSIS AND DISCUSSION OF RESULTS

Anisimov *et al.*[2] have shown that under certain restrictions on the duration and intensity of the ultrashort laser pulse the electron temperature T_e follows without delay the waveform of the laser pulse. A detailed analysis of this question has shown that certain refinements must be introduced in the description of the glow. We therefore analyze these restrictions in greater detail.

It was assumed in Ref. 2 for simplicity that the electronic thermal-conductivity coefficient χ is independent of temperature. In fact, if the electron temperature T_e deviates from the lattice temperature T_i, then χ can have a noticeable dependence on T_e and T_i. Actually, since χ is proportional to the product of T_e by the electron mean free path l, and $l \sim T_i^{-1}$ at lattice temperatures that are high compared with the Debye temperature, the electronic thermal conductivity coefficient is given at $T_e \neq T_i$, $T_i \gg T_D$ by

$$\chi=\chi_0 T_e/T_i. \quad (2)$$

Here χ_0 is the value of the coefficient χ at $T_i = T_e \gg T_D$.

With (2) taken into account, the system of equations used in Ref. 2 for T_e and T_i takes the form

$$\gamma T_e\frac{\partial T_e}{\partial t}=\chi_0\,\mathrm{div}\left(\frac{T_e}{T_i}\,\mathrm{grad}\,T_e\right)-\alpha(T_e-T_i)+q(\mathbf{r},t),$$
$$c_i\frac{\partial T_i}{\partial t}=\alpha(T_e-T_i). \quad (3)$$

Here χ_0 is the linear coefficient of the electronic specific heat, c_i is the specific heat of the lattice and is independent of temperature at $T_i \gg T_D$, and α is a coefficient that characterizes the energy relaxation between the electrons and the lattice, and is shown in a number of papers (see Ref. 2 and the bibliography therein) to be independent of T_e and T_i at $T_i \gg T_D$; $q(\mathbf{r}, t)$ is the laser power absorbed by the metal per unit volume.

It follows from the system (3) that the electron temperature $T(\mathbf{r}, t)$ can be expressed in terms of the laser power at the same instant of time t, i.e., it is inertialess if the following inequalities are satisfied[1)]:

$$\gamma T_e/\alpha \ll \tau, \quad (4)$$
$$c_i/\alpha \gg \tau, \quad (5)$$
$$\frac{\alpha\tau}{c_i}\left(\frac{T_e-T_0}{T_0}\right)\ll 1. \quad (6)$$

Here T_0 is the initial temperature of the metal and τ is the duration of the laser pulse.

If inequalities (4)–(6) are satisfied, the equation for the electron temperature takes the form

$$\frac{\chi_0}{T_0}\,\mathrm{div}\,(T_e\,\mathrm{grad}\,T_e)-\alpha(T_e-T_0)+q(\mathbf{r},t)=0. \quad (7)$$

At $T_e - T_0 \ll T_0$ the problem reduces to the case investigated in Ref. 2. The inequality (6) is then the consequence of the condition (5).

We consider now the opposite and apparently more typical limiting case, when the electron heating temperature is much higher than the initial metal temperature, $T_e(\mathbf{r}, t) \gg T_0$. Assuming that the diameter of the focal spot is much larger than the depth of heating of the electron subsystem, Eq. (7) reduces, as does its counterpart in Ref. 2, to a one-dimensional one. Depending on the relation between the metal-plate thickness d, the heating depth $l_T \sim (\chi_0 T_e/\alpha T_0)^{1/2}$, and the light absorption length $\varkappa^{-1}$, the solution of (7) takes the form

$$T_e(x,t)=\left(\frac{3T_0}{2\alpha\chi_0}\tilde q^2(t)\right)^{1/3}\left[1-x\left(\frac{\alpha^2T_0}{18\chi_0\tilde q(t)}\right)^{1/3}\right]^2,$$
$$d\gg l_T\sim\left(\frac{\alpha^2T_0}{\chi_0\tilde q}\right)^{-1/3}\gg\varkappa^{-1}, \quad (8)$$

$$T_e(x,t)=\tilde q(t)/\alpha d, \quad l_T\sim(\chi_0\tilde q/\alpha^2T_0d)^{1/3}\gg d,$$
$$T_e(x,t)=q(x,t)/\alpha, \quad \varkappa^{-1}\gg l_T\approx(\chi_0 q/\alpha^2T_0)^{1/3}.$$

Here

$$\tilde q(t)=\int_0^d q(x,t)\,dx,$$

and x is the distance from the surface on which the light is incident. We recall that solution (8) is valid subject to satisfaction of the inequalities (4)–(6) and of the additional condition $T_e - T_0 \gg T_0$ at times $0 < t < \tau$. We note that for the metals used in our experiments the heating thickness and the absorption length $\varkappa^{-1}$ are of the same order, and in this case no solution for the electron temperature, corresponding to the first equation in (8), is realized. The lattice temperature under the same conditions is given by an equation that follows from the second equation in (3):

$$T_i(x,t)\approx T_0+\frac{\alpha}{c_i}\int_0^t dt'\,T_e(x,t'),\quad 0<t\ll\frac{c_i}{\alpha}. \quad (9)$$

After the end of the action of the laser pulse ($t > \tau$) the electron temperature drops rapidly, within a time interval $\sim \gamma T_e/\alpha \ll \tau$, to values close to the lattice tem-

perature, which in turn, in view of the inequality (6), differs little from the initial temperature $T_0(T_e - T_i \sim \alpha\tau T_0/c_i)$. The subsequent evolution of the temperatures $T_e \approx T_i$ is determined by the usual single-temperature equation for heat conduction without a source.

We note that the formula (9) for the lattice temperature, which expresses T_i in terms of the electron temperature T_e calls only for satisfaction of condition (5), from which it follows, in particular, that at $0 < t < \tau$ we have $T_i - T_0 \ll T_e - T_0$. As for the inequality (6), while it is mandatory from the point of view that the thermal response be inertialess, as are also (4) and (5), the behavior of the electron temperature at $t > \tau$, when (4) and (5) are satisfied but (6) is not, agrees qualitatively with the case considered above, i.e., at

$$\frac{\gamma T_e}{\alpha} \ll t-\tau \ll \frac{c_i}{\alpha}, \quad T_e - T_i \sim \frac{\alpha\tau}{c_i} T_i,$$

when all the inequalities (4)–(6) hold. To determine the duration of the heating laser pulse by measuring the temperature response of the electrons it suffices to satisfy only the two conditions (4) and (5). On the other hand to reconstruct the pulse waveform it is necessary, generally speaking to satisfy all the inequalities (4)–(6).

We note that the role of the condition (6), which is equivalent to the requirement that the change of the lattice temperature be small $(T_i - T_0 \ll T_0)$, is also to cause the absorbed power $q(x, t)$, which depends on T_i, to follow without inertia the incident laser radiation flux density. For this very reason the quantity R in Eq. (1) is practically independent of time if (6) is satisfied, so that the conditions under which the intensity of the glow of the hot electrons follows without inertia the laser pulse coincide with the conditions considered here, that the electron temperature be inertialess.

To reconstruct the waveform of the laser pulse it is important to have a simple analytic connection between T_e and $q(x, t)$. From this point of view, at $l_T \sim \varkappa^{-1}$, the objects of interest are thin metallic films in which the distribution of the electron temperature $T_e(x, t)$ over the thickness is uniform in first-order approximation [see (8)], and the connection between T_e and q is linear to the same approximation:

$$T_e^0 = \tilde{q}(t)/\alpha d. \tag{10}$$

According to the second equation of (3), the lattice temperature is

$$T_i^0 = Q(t)/c_i d + T_0, \quad Q(t) \equiv \int_0^t dt'\, \tilde{q}(t'). \tag{11}$$

Here $Q(t)$ is the laser-pulse energy absorbed by a unit area up to the instant of time t.

Linearizing Eq. (7) with respect to a small deviation from the solution (10), we obtain an expression for the first-order correction

$$\frac{\partial^2 T_e^1}{\partial x^2} - \frac{T_e^1}{l_T^2} + \frac{1}{\alpha l_T^2}\left[q(x,t) - \frac{\tilde{q}(t)}{d}\right] = 0. \tag{12}$$

The spatial dependence of the function $q(x, t)$ at $\varkappa d \gg 1$ is of the form $\exp(-\varkappa x)$. At $\varkappa d \leq 1$ it is modified to show a smoother behavior of $q(x, t)$. With an aim at obtaining an upper-bound estimate for T_e^1, we retain when solving (12) the $q(x, t) \sim e^{-\varkappa x}$ dependence also for $\varkappa d \leq 1$. Taking into account the boundary condition $\partial T_e^1/\partial x|_{x=0,d} = 0$ we obtain to in first order in the small parameter $(d/l_T)^2$ from (12):

$$T_e^1(x,t) = T_e^0 \left(\frac{d}{l_T}\right)^2 \left(\frac{1}{\varkappa^2 d^2} + \frac{x^2}{2d^2} - \frac{1}{6} + \frac{1/2 - x/d - e^{-\varkappa x}/\varkappa d}{1 - e^{-\varkappa d}}\right). \tag{13}$$

The mean squared relative deviation of the electron temperature from the uniform distribution $T_e^0(t)$ is of the form

$$\frac{1}{d}\left[\int_0^d dx \left(\frac{T_e^1(x,t)}{T_e^0(t)}\right)^2\right]^{1/2} = \left(\frac{d}{l_T}\right)^2 \varphi_0(\varkappa d). \tag{14}$$

The function $\varphi_0(\varkappa d)$ calculated with the aid of (13) takes on for $\varkappa d = \frac{1}{2}$, 1, and 2 the values

$$\varphi_0(1/2) \approx 0.015, \quad \varphi_0(1) \approx 0.028, \quad \varphi_0(2) \approx 0.054.$$

Recognizing that $\varkappa^{-1} \leq l_T$, we arrive at the conclusion that at a plate thickness $d \approx l_T/2$ the deviation of the electron or ion temperature from a uniform distribution is ~1%.

3. MEASUREMENT OF THE CONSTANT α OF THE ENERGY RELAXATION BETWEEN THE ELECTRONS AND THE LATTICE

The inertialess glow induced in metals by picosecond laser pulses can serve as the basis for a method of directly studying the relaxation kinetics of the electron and phonon subsystems in metals. In particular, the essential parameter in the investigation of the electron and lattice relaxation kinetics in metals is the coefficient α. In a number of papers (see Ref. 2 and the bibliography therein) the coefficient α is estimated from data on the electric conductivity, accurate to order of magnitude. A more accurate measurement of α using the electric conductivity is impossible. Knowledge of the exact value of α is very important also in our case for the determination of the limits within which the electron temperature is inertialess in various metals.

From (1) and (11) we easily obtain for α the expression

$$\alpha = \frac{d^{-1}\,\partial Q(t)/\partial t}{T_e(t) - T_0}. \tag{15}$$

We see therefore that to determine α we must measure $T_e(t)$ and $Q(t)$. If the laser pulse is nearly rectangular, then (15) takes the form

$$\alpha = \frac{Q/\tau d}{T_e - T_0}, \quad Q \equiv Q(\tau) = \int_0^\tau \tilde{q}(t)\,dt. \tag{16}$$

Here Q is the total absorbed laser-pulse energy per unit area, and $T_e \equiv T_e(\tau)$ is the electron temperature at the instant of pulse maximum.

By way of illustration we can estimate α approximately from our data (it is not the purpose of this paper to calculate α exactly), by substituting $T_e = 3.5 \cdot 10^3$ K, $\tau = 20$ psec, $Q/d \approx 2.5 \cdot 10^{10}$ erg/cm^3, namely, $\alpha \approx 6 \cdot 10^{17}$ erg/deg · cm^3 · sec. Electric-conductivity data yield $\alpha \sim 10^{17}$–10^{18} erg/deg · cm^3 · sec.

We emphasize that for an exact measurement of α we must use a single laser pulse. Such measurements are presently underway. The methodological error in the measurement of α is determined as shown above and can amount to less than several percent, i.e., the main measurement error is determined by the instrumental errors.

An exact measurement of α for a given material can be of practical importance for the following reasons. Knowing the constant α, we can use (16) to determine the duration of a rectangular laser pulse (or estimate the duration if the pulse is close to rectangular):

$$\tau = \frac{Q/\alpha d}{T_e - T_0}. \qquad (17)$$

It is essential here to measure the maximum value of the electron temperature T_e and the total absorbed energy Q.

Thus, to estimate the pulse duration there is no need for direct measurements with the streak camera. The proposed method of measuring the duration of ultrashort pulses, while subject to the shortcomings inherent in indirect measurement methods (e.g., the impossibility of resolving a fine temporal structure), has the important advantage that it can determine the durations of ultrashort laser pulses in the near and far infrared.

4. CONCLUSION

Inertialess glow of metals makes it possible to observe the onset of hot electrons, and can serve as the basis of a method for studying the properties of metals and for the conversion of IR laser radiation into visible light. This conversion offers prospects of expanding the spectral range of modern photoelectric recorders. This phenomenon is presently under study for various metals under the action of a single picosecond laser pulse. The purpose of further research is to determine the exact range in which the phenomenon is inertialess, to measure the constant α of various metals, to search for means of further expanding the power and laser-pulse-duration ranges of the observed phenomenon. For practical applications, this phenomenon is being investigated for the study of the properties of metals and for the measurement of the temporal parameters of picosecond IR laser pulses.

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[1)]The inequality (6), which is not contained in Ref. 2, corresponds to the condition of weak heating of the lattice, i.e., $T_l - T_0 \ll T_0$. It is due to the dependence of the coefficient of electronic thermal conductivity and of the quantity $q(\mathbf{r},t)$ on the lattice temperature, which is by itself subject to inertia. We note also that the constant α, the absorption coefficient , and the reflection coefficient R are practically independent of time and correspond to their values at $T_e = T_i = T_0$, inasmuch as thanks to (6) we have $T_i \approx T_0$, while the electron heating leads to relative corrections $\sim (T_e/T_F)^2 \ll 1$ (T_F is the Fermi temperature).

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Determination of the mean square displacement of the atomic vibrations in myoglobin molecules by measuring Rayleigh scattering of the Mössbauer radiation

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Rayleigh scattering of Mössbauer radiation (RSMR) is employed to determine the mean square displacement $\langle x^2 \rangle$ of the oscillations of atoms in myoglobin molecules in the crystal state. A procedure is described which has been developed to separate $\langle x^2 \rangle$ of the Mb molecules in a crystalline sample consisting of a mixture of buffer-solution molecules and myoglobin molecules. The experimental value of $\langle x^2 \rangle$ obtained is found to exceed considerably that found by x-ray analysis. It is concluded that determination of $\langle x^2 \rangle$ by the latter method is greatly affected by thermal diffusive scattering, which appreciably lowers the value compared to the true value as determined by the RSMR technique.

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The investigation of the dynamic properties of proteins takes on great importance in connection with the strong influence of these properties on the functional activity of proteins.[1] One promising method for the study of protein dynamics is x-ray diffraction, which yields a map of the mean square displacements $\langle x^2 \rangle$ of the atoms in the molecules under study.[2,3] However, the time scale in this technique is $\tau_x \sim 10^{-15}$ sec, and consequently this method is insensitive to conformational transition frequencies. Static disorder in the arrangement of the molecules in the crystals gives the same contribution to $\langle x^2 \rangle$ as real motions, which may be rather slow ($\sim 10^7$–10^{10} Hz) for protein molecules. Moreover, with an energy resolution to several electron-volts in protein x-ray diffraction, it is difficult to separate elastic scattering from inelastic thermal dif-

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