# *EMF PRODUCED IN SHOCK COMPRESSION OF LANTHANIDES*

V. N. MINEEV, A. G. IVANOV, Yu. V. LISITSYN, E. Z. NOVITSKIĬ and Yu. N. TYUNYAEV

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An emf is observed on shock compression of cerium, europium, and ytterbium samples and is investigated. Possible sources of the emf are considered: a) shock polarization, b) diffusion of carriers from the shock wave front, c) inertial jump-through of carriers, d) thermoelectric power. Some proofs are presented that the emf observed in some cases is due to mechanism a) or b).

PROGRESS in the theory of the electric signals produced upon shock compression of conducting materials is hindered by the lack of experimental data on the separation of the roles of static and dynamic emf sources in explosion experiments. In some papers on this subject[1-3] the effect is associated with the contact between two shock-compressed metals (thermoelectric power). It is noted that the registered values of the emf (~10 mV) exceed by 2–4 times the thermoelectric-power values corresponding to the calculated temperatures of shock-compressed metals in contact, for example copper and nickel. The Seebeck coefficient, obtained from static measurement, was used in calculations. On the other hand, emf's on the order of 0.1–1 V were observed in shock compression of bismuth[4] and silicon of different conductivity[5,6], in which the main effect is associated by the authors with the front of the shock wave (SW), in spite of the fact that the Seebeck coefficient of bismuth and silicon under normal conditions, just like the registered emf, is 10–100 times larger than for the metals investigated in[1-3].

We attempt in the present paper to ascertain the main cause of the emf produced by shock loading of lanthanides. The experimental data were obtained for cerium, samarium, europium, and ytterbium, the resistivities of which under normal conditions are $\rho = (3-9) \times 10^{-5}$ Ω-cm. According to Hall-constant measurements, the conductivity of cerium, europium, and ytterbium is due to positively charged carriers[7]. The electronic structures of these metals differ only in the number of the electrons in the 4f states. For this reason, the physicochemical properties of these elements, with the exception of the magnetic properties, which are determined by the structure of the 4f layer, are close to one another. Under normal conditions, cerium and ytterbium have fcc, europium bcc, and samarium rhombohedral lattices. According to[8], the 4f electrons in these metals are localized at the lattice sites, and the 6s electrons are fully collectivized.

Shock loading realigns the electronic structure of the investigated metals, namely, in the case of europium, ytterbium, and samarium the 6s electrons shift apparently to the d levels of the fifth layer[9,10], and in the case of cerium the electrons move from the 4f band to the 5d band[11]. This is observed for europium at a SW pressure p = 380 kbar, for ytterbium at p = 490 kbar, and for samarium at p = 540 kbar[9,10]. In the case of cerium, the transition takes place at a hydrostatic pressure of 7 kbar[11]. According to the results of quantum-mechanical calculations[12], the electronic transition in metals at high densities and temperatures is accompanied by a jump of the thermodynamic and kinetic electronic characteristics of the metal, particularly the electric conductivity and the form of the Fermi surface, and consequently by a sharp change in the Seebeck coefficient. An experiment[11] performed on cerium has shown that, in accord with the theory, the electronic transition is an isomorphic first-order phase transition (with a jump in volume). This means that within the framework of the theory of free electrons there should be observed a jumplike change of the Fermi energy and, as a consequence, a sharp change in the Seebeck coefficient with changing pressure.

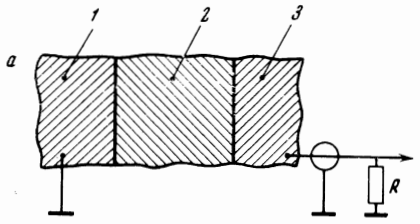


FIG. 1. Experimental setup: 1–screen, 2–sample, 3–electrode.

Thus, if the electric signals produced by shock compression of lanthanides are due to the thermoelectric power, then at pressures corresponding to the electronic transitions we should expect a sharp change in the emf.

## 1. EXPERIMENTS

All the experiments were performed with polycrystalline samples of 1–2 cm diameter and thickness $l$ = 0.185–0.52 cm, containing not less than 99.9% of the main metal. The geometry of the explosive devices[13] and of the samples ensured uniform and constant flow of matter behind the SW front. The measuring circuit (Fig. 1) consisted of a grounded screen 1, an electrode 3, the sample 2, a high-frequency cable, and a load resistance (the input resistance of the oscilloscope was R = 91 Ω). The SW propagated from the screen to the electrode. The acoustic rigidities of the investigated metals and of the aluminum from which the screen and electrode were made differed by no more than 30%. In the calculation of the SW parameters in the samples, we used the dynamic adiabats of the investigated metals[9,10].

Oscillograms of U(t) (the voltage across R) are shown in Fig. 2. An analysis of the oscillograms showed that U(t) arises at the instant when the SW arrives at the boundary between the screen and the sample. The instant

Initial data and results of experiments

| Experiment* No. | l, cm | p, kbar | $t_k$**, μsec | $U_k$, mV | τ, μsec | η, μsec |
|---|---|---|---|---|---|---|
| | | | Ytterbium | | | |
| 1 | 0.395 | 120 | 1.7 | −134 | ~1 | ~1 |
| 2 | 0.293 | 120 | 1.2 | −40 | | |
| 3 | 0.270 | 200 | 1.0 | −370 | | |
| 4 | 0.295 | 200 | 1.1 | −280 | | |
| 5 | 0.296 | 380 | 0.75 | −160 | | |
| 6 | 0.220 | 380 | 0.70 | −135 | | |
| 7 | 0.266 | 780 | 0.70 | 130 | | |
| | | | Europium | | | |
| 8 | 0.185 | 170 | 0.70 | 66 | ⩽0,05 | 0,03 |
| 9 | 0.185 | 170 | 0.70 | 73 | | |
| 10 | 0.520 | 170 | 1.98 | 43 | | 0,15 |
| 11 | 0.185 | 320 | 0.57 | 170 | | |
| 12 | 0.200 | 670 | 0.44 | 150 | | |
| | | | Cerium | | | |
| 13 | 0.430 | 120 | 1.56 | 15 | | |
| 14 | 0.480 | 210 | 1.62 | 15 | | |
| 15 | 0.386 | 390 | 1.07 | −15 | | |

*The experiment number corresponds to the number of the oscillogram on Fig. 2.

**The actual values are given, and differ from the calculated ones by not more than 10%.

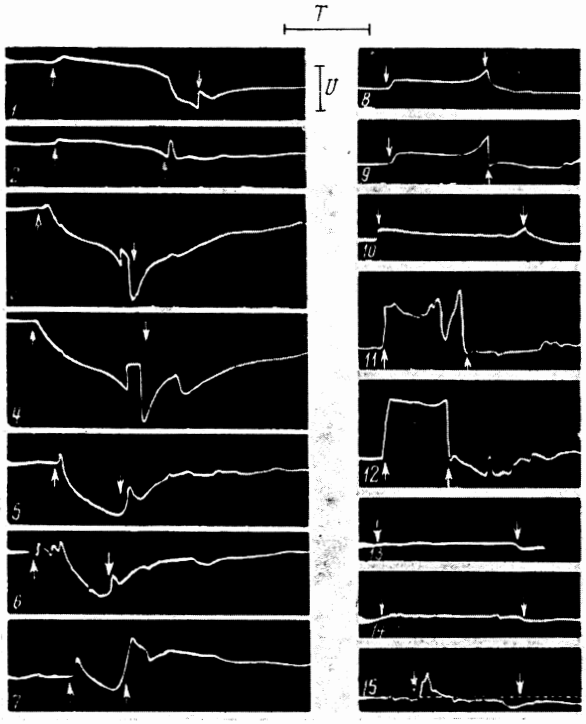


FIG. 2. Oscillograms: 1–7 ytterbium, 8–12 europium, 13–15–cerium, 1, 7, 10, 13–15–T = 1 μsec; 8, 9, 11, 12–T = 2 μsec, U = 150 mV. The arrows on the oscillograms denote the instants of entrance and exit of the SW front into and from the sample. The sweep is from left to right.

when the SW leaves the boundary between the sample and the electrode ($t_c = l/D$, where D is the velocity of the SW front) corresponds in all cases to an inflection of U(t).

Assuming that under conditions of shock compression the value of $\rho$ of the lanthanides does not increase by more than 4–5 orders of magnitude, we get $4l\rho/\pi\Phi^2 \ll R$. Therefore U(t) is equal to the emf induced from the measuring circuit. Favoring this assumption is the fact that under conditions of static compression up to 300 kbar, the change of $\rho(p)$ for ytterbium amounts to only one order of magnitude[14]. The values $U_c = U(t_c)$ in the investigated range of pressures and the characteristic times, namely the growth of the signal upon entry of the SW into the sample ($\tau$) and the decrease of U(t) when $t > t_c$ ($\eta$) are indicated in the table. Experiments performed with samarium samples ($l$ = 0.12–0.19 cm) at p = 140 and 360 kbar have shown that the possible value of the emf in this metal does not exceed the sensitivity threshold of the measuring circuit (< 10 mV).

A characteristic feature of all the obtained plots of U(t) for europium at p = 170 kbar (oscillograms 8–10 on Fig. 2) is the noticeable growth of the voltage at 0.12–0.15 μsec prior to the exit of the SW front from the sample, regardless of the sample thickness.

## 2. DISCUSSION

Let us turn to the experimental setup of Fig. 1, which is described in detail in[6]. If a shock wave moves through the investigated sample, then either a shock wave propagates through the circuit elements in a direction opposite to that of the circuit, or the first shock wave is followed by a rarefaction wave. The former case was realized in the scheme of Fig. 1. From the point of view of the possibility of occurrence of an emf in the circuit, the SW front has a certain distinguishing feature, owing to the dynamic character of the process. Therefore, on a par with the "static" emf sources (the internal contact potential difference), there can occur in the circuit of Fig. 1 dynamic sources due, for example, to effects of dragging of the carriers by the deformed lattice[15,18], deformation of the surface double electric layers in the SW[19], jump-through of the carriers by inertia, and shock polarization.

### Static Emf Sources

We estimate them under the assumption that the Fermi energy $\mu$ of the carriers is determined from the theory of the free-electron gas. Then, at absolute-zero temperature $T_0$ we have $\mu_0 = An^{2/3}$ and for $T_0 < T < T_f$ ($T_f$ is the degeneracy temperature), we have $\mu \approx \mu_0[1 - B(T/\mu_0)^2]$, where $A = (h^2/2m)(3/8\pi)^{2/3}$ and $B = \pi^2 k_1^2/12$; n is the carrier density, m the carrier mass, h Planck's constant, and $k_1$ the Boltzmann con-

stant. The internal contact potential difference is $U = g^{-1}\Delta\mu$, where g is the carrier charge, $\Delta\mu$ the difference between the Fermi energies of the two metals, which differ either in the values of $\mu_0$ (different metals) or in their state (n, T). The total emf of the circuit is $\Sigma U = g^{-1}\Sigma\Delta\mu$.

Such an interpretation of the experimental setup shows that

$$\Sigma U = \frac{B}{g}\left\{T_i^2\left(\frac{1}{\mu_{20}}-\frac{1}{\mu_{10}}\right)+T_1^2\left(\frac{1}{\mu_{10}}-\frac{1}{\mu_{10}\delta_1^{2/3}}\right)\right.$$
$$\left.+T_2^2\left(\frac{1}{\mu_{10}\delta_1^{2/3}}-\frac{1}{\mu_{20}\delta^{2/3}}\right)+T_3^2\left(\frac{1}{\mu_{20}\delta^{2/3}}-\frac{1}{\mu_{20}}\right)\right\},$$

where $\mu_{10}$ and $\mu_{20}$ are the Fermi energies of the carriers in the metals from which the screen and sample are made; $\delta$ and $\delta_1$ are the compressions behind the SW front in the sample and in the screen, $T_i$, $T_1$, $T_2$, and $T_3$ are respectively the initial temperature and the temperatures on the SW front in the sample, on the boundary of the shock-compressed screen and the sample[6], and on the SW front in the screen. The first and third terms of the right-hand side of the obtained expression describe the emf of the pressure thermocouple, which was investigated in detail by Bridgman[20]. The second and fourth terms correspond to the emf due to the difference between the Fermi energies on both sides of the SW in the screen and the sample.

For aluminum $\mu_{10} \approx 10$ eV, and for the investigated lanthanides $\mu_{20} \approx 3$ eV. Putting by way of an estimate $\delta = \delta_1 \approx 2$, $T_i = 300^\circ$ K, and $T_1 = T_2 = T_3 \approx 1000^\circ$ K, we obtain $\Sigma U \sim 1$–2 mV. The contributions of the different terms to $\Sigma U$ are in this case of the same order. The experimentally registered emf is larger by one or two orders of magnitude than $\Sigma U$.

It should be noted, however, that the shapes and lifetimes of the signals can correspond in the cases of europium (Fig. 2, oscillograms 11 and 12) and cerium (Fig. 2, 13 and 14) to the contact emf due to the heating of the contact surfaces between the screen and the sample and between the sample and the electrode by the shock wave. The anomalously large value of the observed signals, according to[15], can be attributed to the high nonequilibrium temperature of the electron gas. On the basis of these considerations, a "contact" nature of the effect, at least for europium (p = 320 and 670 kbar) and cerium (p = 120 and 210 kbar) cannot be excluded.

Nonetheless, the following additional arguments allow us to exclude from consideration static emf sources for ytterbium (p = 120, 200, 380, 780 kbar), europium (p = 170 kbar) and cerium (p = 390 kbar).

1. An analysis of the dependences of the emf on p for ytterbium and europium has shown that in the region of the electronic transition there is observed for ytterbium a reversal in the sign of $U_c$. In the case of europium, the emf does not reverse sign and its value remains approximately constant. For this reason, it is impossible to relate uniquely the observed emf with the change of the electronic structure of ytterbium and europium, and by the same token with the contact emf.

2. The times $\tau$ and $\eta$ are in most cases much larger than the times of establishment of equilibrium of the contact electric processes: $\nu = a^2c/\pi k$[21,22], where c is the specific heat per unit volume, k the thermal conductivity of the substance, and a the characteristic dimension of the region in which the contact electric phenomenon under consideration takes place. Even at $a \sim 100$ Å, we have $\nu = 10^{-13}$ sec (for the investigated lanthanides, $c = 10^{-2}$ cal-cm$^{-3}$deg$^{-1}$ and $k = 3 \times 10^{-2}$ cal-sec$^{-1}$ cm$^{-1}$deg$^{-1}$[23]).

3. During the course of motion of the SW front through the ytterbium samples (p = 120 and 780 kbar), a change takes place not only in the amplitude but also in the sign of U(t). If such a U(t) dependence were to be attributed to the contact emf, this would be evidence of a complex time dependence of the expression $(\mu_1 - \mu_2)/g$, where $\mu_1$ and $\mu_2$ are the Fermi energies of the metals in contact, aluminum and ytterbium. For the reason given in Item 2, a reversal of the sign of the expression $(\mu_1 - \mu_2)/g$ within a time $t_c \sim 1$ $\mu$sec is highly improbable.

### Dynamic Emf Sources

1. Deformation of the surface double electric layer on the screen-sample contact in the SW can lead to a current that exists for a time $\Delta/D \sim 10^{-11}$ sec ($\Delta \sim 10^{-6}$ cm is the width of the electric double layer). Since this time interval is smaller by several orders of magnitude, than the time resolution of the measuring circuit, this effect could not be registered, even if the influence of the skewing of the SW front on U(t) is taken into account[24].

2. The dragging effect. This phenomenon is similar to the acoustoelectric effect[25] and consists of dynamic electron-phonon interaction, which leads to dragging of the carriers by the phonons polarized in the direction of SW motion. Coleburn, Solow, and Wiley[17] proposed that the occurrence of the acoustoelectric effect during the process of shock compression of a conductor should lead to the diffusion of the carriers from the SW front. They attribute to this phenomenon the registered glow of a luminor on the free surface of an aluminum plate 0.63 $\mu$sec prior to the instant of emergence of a SW front from the plate (p = 270 kbar). The diffusion of the carriers from the SW front was observed also in bismuth[4]. The noticeable growth of U(t) upon approach of the SW to the electrode (independently of $l$) is a characteristic of this metal. A similar phenomenon is observed in the case of europium (p = 170 kbar). The growth of U(t) upon approach of the SW front to the electrode can be attributed to the penetration of the "precursor" charge ~0.04 cm into the unperturbed substance. We emphasize that whereas in the case of bismuth the carriers diffusing from the SW front have a negative charge, in the case of europium they are positive. Consequently, the sign of the charges diffusing from the SW front coincides in both cases with the sign of the majority carriers under normal conditions. Thus, the occurrence of the emf due to carrier diffusion on the front of the shock wave is likely.

3. The emf on the SW front produced by jumping through of the carriers by inertia is connected with the jumplike change in the velocity of the matter in the SW front (V), as a result of which the electroneutrality is violated and an electric force (gE) is produced and opposes such a jump (a phenomenon analogous to the Mandel'shtam-Papaleksi-Tolman effect—the MPT effect). From the condition that the forces be equal

we get $gE = mV/\omega$, where $\omega$ is the time of deceleration of carriers with mass m. This expression coincides with the formula for estimation of the MPT effect[26]. A specific feature of this phenomenon in SW, in our opinion, is that the field E can penetrate only into the region of the shock transition (with width $\lambda$) and can exist in stationary manner as the SW propagates through the sample. Then the emf is $U = E\lambda = mV\lambda/g\omega$. We put $\lambda = \omega D$, and then $U = mVD/g$. Assuming that the carriers are electrons and putting $V \sim 10^5$ cm/sec and $D \sim 5 \times 10^5$ cm/sec, we get $U \sim 10^{-5}$ V (a voltage pulse $Ut_c \sim 10^{-11}$ V-sec). Such signals could not be measured in our experiments. For comparison we note that the MPT effect is characterized by a value $\sim 10^{-6}$ V-sec[26].

It is obvious that whereas in the case of Item 2 the sign of the emf on the SW front coincides with the sign of the carriers, in the case of Item 3 the sign is opposite.

4. The shock polarization of the investigated metals can have an ionic and (or) electronic nature. However, the large times $\tau$ and $\eta$ in comparison with the relaxation time of the electron polarization ($10^{-13}$—$10^{-14}$ sec) make it possible either to exclude the electronic polarization from consideration, or to admit the formation behind the SW front of oriented trap-electron systems with time of mechanical[6] relaxation $\sim 10^{-6}$ sec (polarization of the Maxwell-Wagner type in dielectrics[27]).

Ionic polarization of conducting materials can be due, for example, to the formation and orientation in the SW front of charged vacancy plus interstitial ion pairs. In this case the current flowing in the external circuit is the result of the interaction of the carriers with the indicated pairs during the process of their recombination. A temporal characteristic of the recombination process of Frenkel pairs in a metal can in first approximation be assumed to be the average time between the jumps of the ions, which lies in the interval from $\sim 10$ sec (under normal conditions) to $\sim 10^{-11}$ sec (upon melting). If the characteristic time of the recombination process of the charged Frenkel pairs is much larger than the carrier relaxation time, then the law governing the variation of the current in the external circuit will coincide with the law of variation of the polarization behind the SW front. An analogous situation is realized in shock polarization of semiconductors and is considered in detail in[6].

We note that the phenomenological theory of shock polarization in the form developed in[28] cannot be employed here to obtain quantitative data, since in our case certain relations used in the derivation of the theory no longer are valid. We recall that at each point of a conducting medium the electric field E resulting from shock compression of the substance (at any velocity of the SW front) exists for a time on the order of $\theta = \rho\epsilon/4\pi$, where $\epsilon$ is the dielectric constant of the substance. For the substances under consideration, with $\epsilon \sim 1$, the time is $\theta \sim 10^{-17}$—$10^{-18}$ sec. On the other hand, the value of $\theta$ cannot be smaller than the average time interval between two carrier collisions ($\sim 10^{-14}$ sec).

## CONCLUSION

It has been established that the emf produced upon shock compression of ytterbium (p = 120, 200, 380, and 780 kbar), europium (p = 170 kbar) and cerium (p = 390 kbar) is not connected with contact effects, and is due principally to the volume redistribution of the charges in the SW front. In the case of europium (p = 320 and 670 kbar) and cerium (p = 120 and 210 kbar) a role of the contact effects in the observed emf cannot be completely excluded.

The effect of occurrence of an emf on a SW front is characteristic of a large class of substances: dielectrics (ionic crystals[29]), organic compounds[30,31], semiconductors (crystals of silicon[5,6] and germanium[5]) and metals (present communication).

We note that the quantum-mechanical method of calculating the electronic structure of compressed and heated metals that was proposed in[32] permits, in principle, estimation of the conductivity and the Fermi energy of the lanthanides investigated by us. Such calculations and their analysis as applied to the electric effects observed in the present paper will contribute not only to further understanding of the nature of the observed emf, but will provide more exact information on the electronic structure of these metals.

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