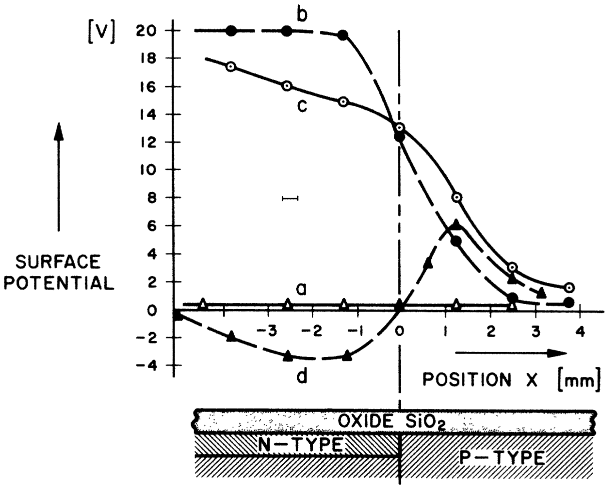

FIG. 1. Surface potential vs gold of a silicon p-n junction covered with approximately 1000 Å of oxide, plotted versus distance x from the junction. Curve a, equilibrium; curve b, reverse bias of +20 V applied to n region ($x < 0$), p region grounded; curve c, after exposure to "wet" ambient with bias applied; curve d, "dry" ambient restored and bias removed; evidence for residual charges on oxide. Bar indicates size of vibrating probe.

plied field causes a motion and redistribution of charges on the oxide surface and possibly also within the oxide. The distribution is such as to reduce the external field, i.e., positive charges accumulate on the p side, which is biased negative with respect to the n side, and vice versa. This charge arrangement is aided by water vapor, which probably increases concentration and mobility of the ions or dipoles. It is remarkable that after voltage removal, the remaining charge corresponds to as much as 50% of the previous bias.

The potential distributions, as shown in curves b to d, and the associated time constants of the drifts depend sensitively upon surface treatment and ambient. For example, a soaking with methanol yielded drift time constants of a few seconds for the positive charges, but of several minutes for the negative charges. Such details are currently being investigated with the hope to identify nature and transport mechanism of the charged entities.

These surface charges may be interpreted as a form of mobile slow states. They, in turn, exert influences upon the underlying semiconductor. We found correlations between surface potential and channel conductance as well as with breakdown voltage. These results are similar to those of Atalla, Bray, and Lindner[3] and in agreement with models[4] ascribing changes in junction behavior to surface charges.

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[1] W. L. Brown, Phys. Rev. 91, 518 (1953).

[2] H. Statz and G. A. deMars, Phys. Rev. 111, 169 (1958).

[3] M. M. Atalla, A. R. Bray, and R. Lindner, Suppl. Proc. Inst. Elec. Engrs. (London) Pt. B 106, 1130 (1959).

[4] C. G. B. Garrett and W. H. Brattain, J. Appl. Phys. 27, 299 (1956).

FERROMAGNETIC RELAXATION IN EUROPIUM IRON GARNET

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In recent years two theories have been proposed to explain ferromagnetic relaxation in rare earth iron garnets (REIG). The earlier theory, proposed by de Gennes et al.,[1,2] is called the "fast" relaxation theory, because the assumption is made that $\tau_{\mathrm{RE}} \ll \omega^{-1}$ over most of the temperature range, where ω is the operating frequency. The other theory, originally proposed by Galt[3] and Clogston[4] and developed in some detail for YbIG by Teale and Tweedale[5] and by Van Vleck and Orbach[6] as well as more generally by Hartmann-Boutron,[7] is called the "slow" relaxation theory because here τ_{RE} is assumed comparable to ω over the temperature range of interest.

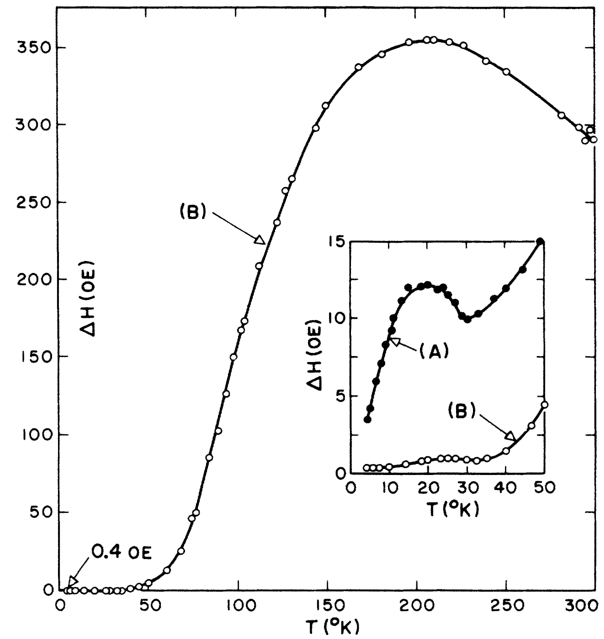

FIG. 1. ΔH vs T on a 1.4-mm sphere of EuIG along the [111] axis at 9980 Mc/sec. Curves (A) and (B) correspond to crystal growth from ordinary and highly purified lead oxide-boron oxide fluxes, respectively.

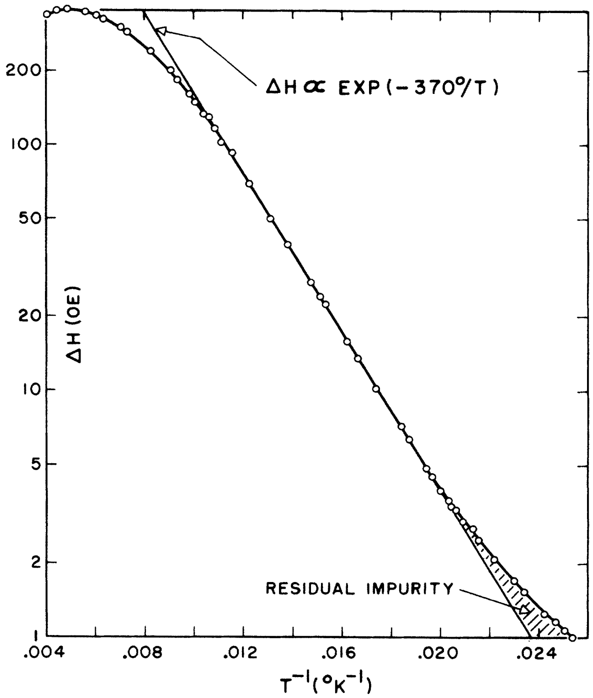

FIG. 2. Plot of $\ln\Delta H$ vs T^{-1} for the data in curve (B) of Fig. 1. In this figure the residual 0.4-Oe linewidth at 4.2°K, probably due to surface imperfections, has been subtracted off. Due to experimental inaccuracies and the residual impurities, the fit to $\exp(-370°\text{K}/T)$ may not be as good as it appears. The data may also be fitted acceptably by $\Delta H \propto T^{-1}\exp(-410°\text{K}/T)$.

An important difference between the predictions of the two theories concerns the frequency dependence of the observed maxima in $\Delta H(T)$. In the "fast" relaxation theory $\Delta H \propto \omega$ over the entire linewidth peak, while in the "slow" relaxation theory $\Delta H \propto \omega$ on the high-temperature side and $\Delta H \propto \omega^{-1}$ on the low-temperature side. The available experimental evidence[5,8] tends to confirm the "slow" relaxation theory, but more quantitative understanding is greatly needed.

In this Letter we should like to report observations of ΔH in EuIG which have the unusual feature that while in agreement with the predictions of the "fast" theory on both sides of the linewidth maximum, they can still nevertheless be explained more plausibly by a "slow" relaxation theory. It is possible that EuIG is unique in this respect.

In Fig. 1 we see $\Delta H(T)$ at 9980 Mc/sec on single crystal spheres on EuIG grown from (A) ordinary and (B) highly purified[9] lead oxide-boron oxide fluxes.[10] We are reasonably certain that the difference at low temperatures is due to the much lower silicon content in the fluxes in case (B). The purity of the Eu_2O_3 in (B) is 99.99%. Figure 2 shows $\ln\Delta H$ vs T^{-1}, and Fig. 3 shows g_{eff} vs T and g_{eff} compared to $g(\text{YIG})M_S(\text{EuIG})/M_S(\text{YIG})$.[11] The measurements were made using spheres free to rotate in a quartz tube, which was spaced off the end wall of a shorted rectangular waveguide. The data can be summa-

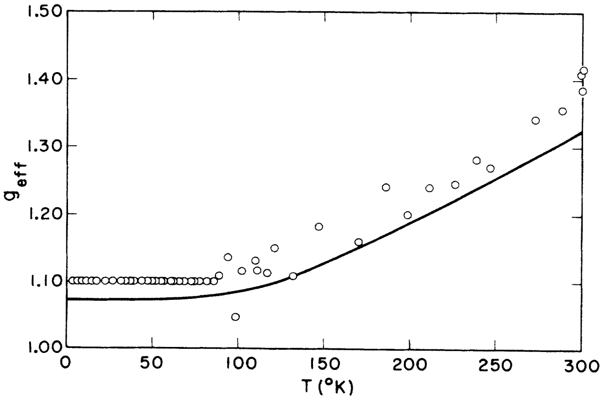

FIG. 3. Values of g_{eff} vs T measured on the sphere used for curve (B) in Fig. 1. The solid curve is $g(\text{YIG}) \times M_S(\text{EuIG})/M_S(\text{YIG})$. The abrupt increase in the scatter of the data at ~85°K is due mainly to the large values of ΔH above this temperature.

rized as follows:

(1) g_{eff} is within a few percent of g(YIG) $\times M_s$(EuIG)$/M_s$(YIG) from 1.5° to 300°K. These data were obtained from measurements of the applied field H_0 for resonance at each temperature for several frequencies between 9600 Mc/sec and 12300 Mc/sec and fitting the results to $\omega=(g_{\text{eff}}e/2mc)(H_0+H_a)$. At 4.2°K, $H_a=4983$ Oe for the effective anisotropy field along [111].

(2) $\Delta H \propto \exp(-370°\text{K}/T)$ from ~50 to ~100°K.

(3) ΔH varies as ω (or a little faster) from 6000 Mc/sec to 18320 Mc/sec,[12] over the range 45 to 300°K.

(4) The temperature for $\Delta H_{\max}$ at 9980 Mc/sec is approximately the same as that observed at 18320 Mc/sec by Miyadai,[12] namely, ~210°K.

Point (1) has been explained satisfactorily by Wolf,[13] who pointed out that because of the $J=0$ ground state, the real part of the effective g factor for Eu^{3+} is very large. This is sufficient to yield the agreement shown in Fig. 3.

Points (2)-(4) can be understood on the basis of the longitudinal (so-called "slow") or, with less probability, the transverse (a "fast") relaxation mechanism. The present "slow" relaxation mechanism, however, differs from the classic "slow" mechanism since, as will be shown below, $\omega\tau \neq 1$ at the linewidth maximum. We note first that any relaxation effects undoubtedly come from the components of the states $J=1$, as the state $J=0$ has only an induced moment, and the transverse relaxation associated with transitions between $J=0$ and $J=1$ is negligible since the difference in energy $\hbar\bar{\omega}$ of the states $J=0$ and $J=1$ is so large. The relaxation time of any of the components into which $J=1$ is split by the exchange and crystalline fields is undoubtedly short. Orbit-lattice interaction between $J=0$ and $J=1$ is forbidden by the triangle rule (except higher order effects associated with the fact that the exchange and crystalline fields make J cease to be completely "a good quantum number"). However, there are two relaxation processes which can make τ short: (a) interaction with iron-lattice magnons resonant to the transitions between $J=0$ and $J=1$, and (b) orbit-lattice interaction between the three components of $J=1$. The relative importance of (a) and (b) is being studied by Mr. D. Huber, and his preliminary results are that (a) predominates, since the iron-lattice magnon spectrum extends well above $\bar{\omega}$. In either case the relaxation time presumably is short, so that the $1+\omega^2\tau^2$ denominator characteristic of the longitudinal mechanism may be replaced by unity. The linewidth is then proportional to ω and the temperature for $\Delta H_{\max}$ is independent of ω, in agreement with points (3) and (4). The important point is that τ^{-1} does not approach zero at low temperatures and instead will vary with temperature as[14]

$$\tau=\tau_0\tanh(\hbar\bar{\omega}/2kT) \tag{1}$$

for process (a). The formula for (b) is similar to (1) except that $\bar{\omega}$ is replaced by ω_{11}, where $\hbar\omega_{11}$ is some appropriate splitting interval internal to $J=1$. (Actually the state $J=1$ is a three- rather than a two-component system, but our discussion is sufficient for qualitative purposes.)

If the polar axis is perpendicular to the applied static field, then Clogston's formula[4] shows that the linewidth caused by longitudinal relaxation for small RE concentration is

$$\Delta\omega=\frac{-e}{2mcM}\sum_{j=1}^{3}\left(\frac{\partial E_j}{\partial\theta}\frac{\partial N_j}{\partial\theta}+\frac{\partial E_j}{\partial\varphi}\frac{\partial N_j}{\partial\varphi}\right)\frac{\omega\tau}{1+\omega^2\tau^2}, \tag{2}$$

where $\Delta\omega=(e/2mc)\Delta H$, M is the magnetization of YIG, and 1, 2, 3 are the three components of $J=1$. The derivatives are to be calculated for infinitesimal deviations of the magnetization of the YIG from its equilibrium direction. If the origin is taken at $J=0$, the population of a component is $N_j=N\exp(-E_j/kT)/[1+\sum\exp(-E_j/kT)]$. From (2) it follows that if the splitting ($\sim\omega_{11}$) of the $J=1$ states is small compared to their separation $\bar{\omega}$ from $J=0$, and if $\omega^2\tau^2 \ll 1$, then

$$\Delta\omega=\frac{C\hbar\omega_{11}^2}{kT}\Phi\omega\tau\frac{\exp(-\hbar\bar{\omega}/kT)}{1+3\exp(-\hbar\bar{\omega}/kT)}, \tag{3}$$

where C is the atomic ratio of Eu to Fe (3/5 for EuIG) and Φ is a dimensionless angular factor which vanishes for all angles only if the state $J=1$ is unsplit by the crystalline field and if, in addition, the exchange coupling is isotropic. More exact formulas than (3) are readily obtained from (2) to allow for the fact that the exponential factors are not the same for all three components of $J=1$.

By combining (3) and (1), the observed qualitative features of the variation of linewidth with temperature are well reproduced, mainly a linewidth vanishing like $e^{-a/T}$ at low T and a maximum at $T=225°\text{K}$, with a free ion value of $\hbar\bar{\omega}/k=480°\text{K}$.

If, instead, the transverse mechanism is assumed to predominate, approximately the same type of variation with temperature as given by (1) and (3) is again obtained, provided $\hbar\omega_{11} \ll kT$ and provided the transverse relaxation time is $\leq 1/\omega_{11} \sim 10^{-13}$ sec, a drastic condition. More detailed calculations will be made to determine the relative importance of the two mechanisms for different assumed splitting patterns and relaxation times. Also further study of the angular dependence is indicated and is under way. At present it is perhaps premature to decide between the two mechanisms, but we are inclined to favor the longitudinal one because there is experimental evidence that the splitting of the $J=1$ levels is quite large.[15] Thus the relaxation time has to be quite short to make the transverse process effective, and too short a relaxation time might imply more diffuse spectral lines than are observed. With the longitudinal model, a value of τ in the range 10^{-11} to 10^{-12} sec at $T=0$ seems to be satisfactory.

We should like to thank L. R. Walker, S. Geller, and J. A. Koningstein for a number of helpful conversations. We also wish to thank E. M. Kelly for help in the crystal growth, and R. C. Petersen and R. Morris for technical assistance.

[1] P.-G. de Gennes, C. Kittel, and A. M. Portis, Phys. Rev. 116, 323 (1959).

[2] C. Kittel, J. Appl. Phys., Suppl. 31, 11S (1960).

[3] J. K. Galt, Bell System Tech. J. 33, 1023 (1954).

[4] A. M. Clogston, Bell System Tech. J. 34, 739 (1955).

[5] R. W. Teale and K. Tweedale, Phys. Letters 1, 298 (1962).

[6] J. H. Van Vleck and R. Orbach, Phys. Rev. Letters 11, 65 (1963).

[7] F. Hartmann-Boutron, Compt. Rend. 256, 4412 (1963); and (to be published).

[8] J. F. Dillon, Jr., Phys. Rev. 127, 1495 (1962).

[9] Eu_2O_3 and flux materials purified by M. M. Woyski, American Potash and Chemical Corp., Whittier, California.

[10] J. P. Remeika (to be published).

[11] The magnetization data for YIG and EuIG are from S. Geller, H. J. Williams, R. C. Sherwood, J. P. Remeika, and G. P. Espinosa, Phys. Rev. 131, 1080 (1963).

[12] The data at 18 320 Mc/sec are from T. Miyadi, J. Phys. Soc. Japan 15, 2205 (1960).

[13] W. P. Wolf, J. Phys. Soc. Japan 15, 2104 (1960). The special situation involved in the g value for ions in $J=0$ states is also implicit in Kittel's exclusion of such ions from consideration in the original theory, C. Kittel, Phys. Rev. 115, 1587 (1959).

[14] R. Orbach, J. Appl. Phys. 33, 2144 (1962).

[15] J. A. Koningstein (to be published).

RAPID ANNIHILATIONS OF POSITRONS IN POLYATOMIC GASES[*]

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This report presents experimental values of the annihilation rates in gases of slow positrons which have failed to form positronium. The observed values for argon, methane, ethane, propane, n-butane, isobutane, and carbon tetrachloride are found to exceed the values derived from Eq. (1) below by factors ranging from 3 to 700.

The mode(s) of annihilation of positrons which fail to form positronium have rarely been investigated. In 1951 Deutsch[1] demonstrated the existence of a component of annihilation in oxygen for which the half-life is inversely proportional to the gas pressure. The inverse proportionality is exactly what is expected of the annihilation of free positrons according to the Dirac theory[2] in the low-velocity approximation. The Dirac theory predicts the annihilation rate

$$\lambda = n \times 0.748 \times 10^{-14} \text{ sec}^{-1}, \qquad (1)$$

where n is the number of electrons per cm^3 available for annihilation. This simple formula takes no account of the Coulomb attraction between positrons and electrons. The graph published by Deutsch[1] corresponds to

$$\lambda = n \times 1.6 \times 10^{-14} \text{ sec}^{-1}$$

for oxygen, where we have interpreted n as the number of valence electrons per cm^3 in oxygen. Daniel and Stump[3] have published data for helium which provide a similar result:

$$\lambda/n = 1.2 \times 10^{-14} \text{ cm}^3/\text{sec}.$$

The observed λ was in each case slightly greater than the Dirac value. It is usually supposed that a positron approaching a neutral molecule causes an electric polarization sufficient to account for the enhanced annihilation rate. In what follows we shall continue to use n to represent the number of valence electrons per cm^3, and shall use n_m as the number of molecules per cm^3.