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Faraday rotation near the Curie point of EuO[†]

Cheng-Cher Huang*

Department of Physics and Laboratory for Research on the Structure of Matter, University of Pennsylvania, Philadelphia, Pennsylvania 19174

John T. Ho[‡]

Department of Physics, University of Houston, Houston, Texas 77004

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The static magnetic properties of EuO near the Curie point have been measured using Faraday rotation. Comparison between magnetometer data reveals a small nonlinear term in the relation between rotation and magnetization. With this correction, our results yield critical parameters $\beta = 0.370 \pm 0.006$, $B = 1.10 \pm 0.04$, $\gamma = 1.30 \pm 0.02$, and $\Gamma = 0.397 \pm 0.012$. The value of γ agrees with other static experiments, but disagrees with neutron-scattering measurements and with theory. All the data can be scaled using these exponents, and are consistent with the two proposed parametric equations of state, with coefficients $a_1 = 2.56$, $k_1 = 1.025$ and $a_2 = 1.78$, $k_2 = 0.718$. Our data predict specific-heat parameters $\alpha = -0.04 \pm 0.03$ and $A/A' = 1.31 \pm 0.05$, in agreement with experimental results analyzed with nonsingular correction terms.

I. INTRODUCTION

We have measured the static magnetic properties of EuO near the Curie point using the Faraday effect. The critical behavior of EuO, being a good approximation to an isotropic classical Heisenberg ferromagnet, has been studied extensively.[1-6] An apparent difference has emerged between the results of static[1,4] and neutron scattering[2] measurements. Our experiment was motivated partly by this discrepancy, and partly by the need to provide detailed data in the critical region of EuO for comparison with proposed scaling equations of state, specific-heat measurements, and theoretical calculations.

The Faraday effect has been demonstrated to be a sensitive method of measuring the magnetization of materials which exhibit large magnetic rotations at suitable wavelengths.[7,8] In previous studies on $CrBr_3$ and YIG, the Faraday rotation was found to be directly proportional to the magnetization. In EuO, by comparing our results with magnetometer data,[1] we have found a small nonlinear term in the relation between rotation and magnetization. This unusual nonlinearity is believed to be related to the bandwidth of the excited states being comparable to the absorption band gap. This correction has therefore been incorporated into our data analysis.

Our results yield the critical exponents $\beta = 0.370 \pm 0.006$ and $\gamma = 1.30 \pm 0.02$. The latter supports previous static measurements but disagrees with neutron scattering results. It is also different from theoretical predictions, with or without dipolar interactions. With these values of the exponents, all the data can be scaled into a universal function. We have found that both proposed versions of the parametric equation of state describe the data satisfactorily and obtained values of the relevant coefficients. A preliminary report on part of this work has been presented previously.[9]

II. EXPERIMENTAL

The measurements were made at a wavelength of 3.39 μm, at which we found EuO to be quite transparent and to exhibit a saturation Faraday rotation of 2.4×10^4 rad m^{-1}, in agreement with the results of Dimmock *et al.*[10] The single-crystal samples were obtained from Dr. M. W. Shafer of IBM. They were classified as having class II stoichiometry (with about 1% Eu vacancies) as characterized by infrared and conductivity measurements.[11] Although the data presented here are from one sample, consistent results were obtained using a second sample. The polarimeter, consisting of a He-Ne laser, a Soleil-Babinet compensator, a rotating calcite analyzer, a PbS detector, and a lock-in amplifier, had a sensitivity of 9×10^{-4} rad, corresponding to 5×10^{-5} of the saturation magnetization of the sample. The temperature was controlled to a stability of 10^{-3} K in an optical Dewar and measured with a Pt resistance thermometer. External fields up to 10 kG were obtained in a 15-in. electromagnet and measured with a Hall gaussmeter. Details of the measuring procedure has been described elsewhere.[12]

III. DATA ANALYSIS AND RESULTS

The Faraday rotation θ was first measured as a function of applied field H_a up to 500 G, for 48 isotherms between 69 and 77 K. All the isotherms show a linear relation between θ and H_a, with a slope

$$\phi = \left(\frac{d\theta}{dH_a}\right)_T . \tag{1}$$

The ten isotherms which are below and very close to T_c have the same initial slope ϕ_0 within experimental errors. In the range of magnetization M of these data, the nonlinear effects to be discussed later in this section are negligible, and we can assume the relation[7,13]

$$\theta = AM, \tag{2}$$

where A is a constant. The internal field H is given by

$$H = H_a - DM, \tag{3}$$

where D is the demagnetizing constant. The value of D for our disk-shaped sample is 7.5. The susceptibility

$$\chi = \left(\frac{\partial M}{\partial H}\right)_T \tag{4}$$

above T_c is then given by

$$\chi = \phi / D(\phi_0 - \phi). \tag{5}$$

The quantities M, H, and T are expressed in terms of dimensionless units

$$m = M/M\,(0\text{ K}), \tag{6}$$

$$h = (g\mu_B S/kT_c)H, \tag{7}$$

and

$$t = (T - T_c)/T_c, \tag{8}$$

respectively. For EuO, the values used are $4\pi M$ (0 K) = 2.4×10^4 G and $g\mu_B S/kT_c = 6.82\times10^{-6}$ G^{-1}. A least-squares fit of the susceptibility data to the equation

$$\chi = \Gamma t^{-\gamma} \tag{9}$$

yielded the optimum parameters

$$\gamma = 1.30 \pm 0.02, \tag{10}$$

$$\Gamma = 0.397 \pm 0.012, \tag{11}$$

$$T_c = 69.105 \pm 0.02\text{ K}. \tag{12}$$

As illustrated in Fig. 1, the single exponent describes the divergence with no appreciable systematic deviations in the entire range $0.005 < t < 0.1$ of the measurement.

Having determined the values of γ and T_c, 18 isotherms between 66.683 and 77.261 K were measured from low fields up to 9.9 kG. By comparing our data with the magnetization data of Menyuk, Dwight, and Reed[1] using a vibrating-sample magnetometer, it became obvious the Faraday rotation at sufficiently high magnetization cannot be described by the simple linear relation in Eq. (2). This is not surprising, since Eq. (2) is expected to hold only if the bandwidth of the excited states is small compared to the absorption band gap.[7,13] In EuO, the $4f^6 5d$ excited states have a bandwidth of 0.5 eV, while the absorption edge is at 1.1 eV.[14] The data were therefore analyzed by using the simplest extension to Eq. (2) consistent with symmetry,

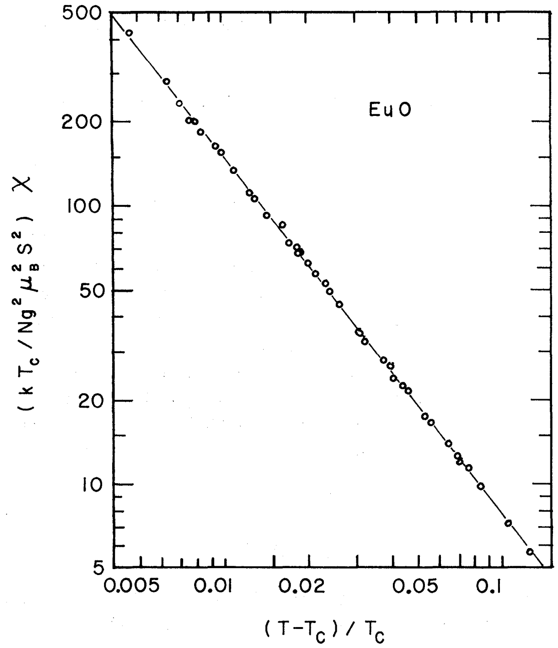

FIG. 1. Temperature dependence of susceptibility χ of EuO above T_c. The line has a slope $\gamma = 1.30$.

$$\theta = A_1 M + A_3 M^3. \tag{13}$$

The coefficients A_1 and A_3 were determined by comparing our data to those of Menyuk *et al.*[1] along the critical isotherm, assuming Eqs. (3) and (13). An optimum value of the demagnetizing constant D was also generated in this fitting procedure, and was found to be within 6% of the value expected from the measured geometry of the sample. This allows us to estimate that any errors in the choice of D due to inhomogeneities in the demagnetizing field, because the sample is not an ellipsoid of revolution, are no more than 6%. The maximum effect of such an uncertainty in D on the data analysis is a shift in the values of the critical amplitudes, as can be seen in Eq. (5). The correction to the data resulting from the introduction of the cubic term in Eq. (13) has a contribution of 15% at the maximum magnetization we measured, and is typically much less. A similar but less-pronounced nonlinear magneto-optical effect has been observed in EuS.[15] Since the Faraday rotation depends on the absorption edge,[7,13] which in EuO is known to exhibit a red shift with decreasing temperature around T_c,[16] we have also included a small temperature dependence in the coefficient A_1. This represents a correction of less than 4% for most of the data.

For each measured rotation, the corresponding M and H were calculated using Eqs. (3) and (13). Figure 2 shows a plot of m^2 vs h/m for nine isotherms. The critical exponent β along the coexistence curve was obtained by extrapolating the data below T_c to zero internal field. The temperature dependence of the spontaneous magnetization m was fitted to the expression

$$m = B(-t)^{\beta}. \tag{14}$$

The resultant parameters are

$$\beta = 0.370 \pm 0.006, \tag{15}$$

$$B = 1.10 \pm 0.04, \tag{16}$$

$$T_c = 69.104 \pm 0.02 \text{ K}. \tag{17}$$

Knowing β and γ, the scaling hypothesis can be checked by plotting $\tilde{h} = h|t|^{-(\beta+\gamma)}$ as a function of $\tilde{m} = m|t|^{-\beta}$ for all the data. As shown in Fig. 3, all the data fall on a universal curve with two branches, one for $t > 0$ and one for $t < 0$. Analyticity of the equation of state requires the following expansion to hold near the critical isochore[17]:

$$\tilde{h} = c_1\tilde{m} + c_3\tilde{m}^3 + c_5\tilde{m}^5 + \cdots, \tag{18}$$

or, equivalently,

$$h = c_1 m t^{\gamma} + c_3 m^3 t^{\gamma-2\beta} + c_5 m^5 t^{\gamma-4\beta} + \cdots . \tag{19}$$

We find the $t > 0$ branch of Fig. 3 up to $\tilde{m} = 2.0$ can be fitted by the first three terms of Eq. (18), with the coefficients $c_1 = 2.49 \pm 0.10$, $c_3 = 2.53 \pm 0.18$, and $c_5 = 0.96 \pm 0.24$.

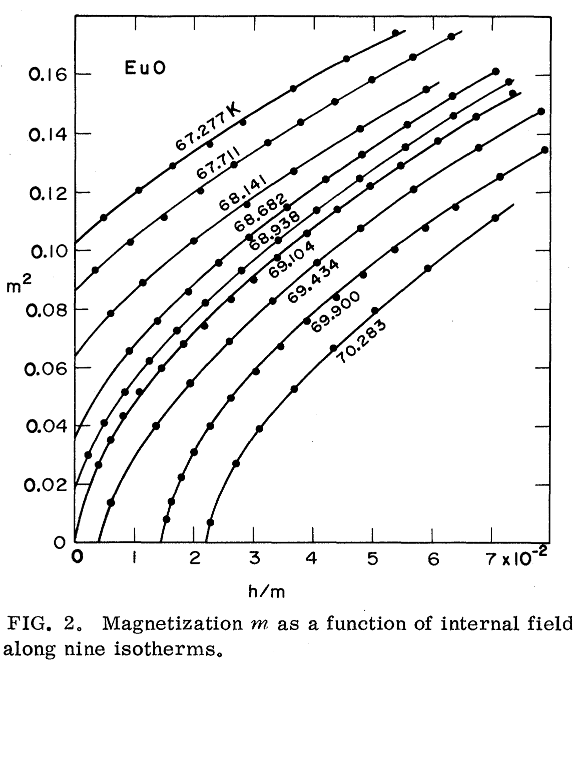

FIG. 2. Magnetization m as a function of internal field h along nine isotherms.

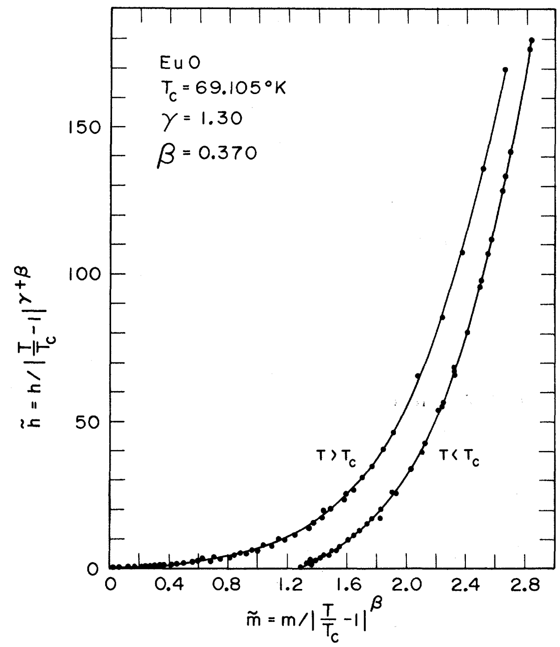

FIG. 3. Universal plot of scaled magnetic field vs scaled magnetization.

IV. PARAMETRIC EQUATIONS OF STATE

We have fitted the data to the parametiric equations of state proposed by Schofield[18,19] and by Ho and Litster.[7] The comparison is more extensive and conclusive than previous attempts,[1,4] and provides values of coefficients that would be useful in future applications in which a concise knowledge of the static critical behavior of EuO is required.

The first parametric equation (PE1), called the linear model, has the form[19]

$$h = a_1\theta(1 - \theta^2) r^{\beta+\gamma}, \tag{20a}$$

$$t = (1 - b_1^2\theta^2) r, \tag{20b}$$

$$m = m(\theta) r^{\beta}, \tag{20c}$$

where

$$m(\theta) = k_1\theta, \tag{20d}$$

$$b_1^2 = (\gamma - 2\beta)/\gamma(1 - 2\beta). \tag{20e}$$

In PE1, the parameter r is a direct measure of the specific heat C_M at constant magnetization. Using the values of β and γ obtained in Sec. III, a least-squares fit of the data to Eq. (20) yielded the optimum coefficients

$$a_1 = 2.56, \tag{21}$$

$$k_1 = 1.025. \tag{22}$$

A comparison of the experimental function $m(\theta)$ with Eq. (20d) is shown in Fig. 4. As a consistency check, we have calculated the coefficients mentioned in Sec. III in terms of the constants obtained in PE1.

The amplitudes along the coexistence curve and the critical isochore are given by

$$B=k_1(b_1^2-1)^{-\beta}, \tag{23}$$

$$\Gamma=k_1/a_1. \tag{24}$$

Using the known values of a_1, k_1, β, and γ, Eqs. (23) and (24) imply $B=1.07$ and $\Gamma=0.400$, in agreement with the values shown in Eqs. (11) and (16). The coefficients in the expansion in Eq. (18) are given by

$$c_1=a_1/k_1, \tag{25}$$

$$c_3=a_1(\gamma b_1^2-1)/k_1^3, \tag{26}$$

$$c_5=a_1 b_1^2[b_1^2\gamma(\gamma-4\beta+1)-2(\gamma-2\beta)]/2k_1^5. \tag{27}$$

The resultant values $c_1=2.50$, $c_3=2.56$, and $c_5=1.05$ are also consistent with the results obtained by direct fitting to Eq. (18).

The second parametric equation (PE2) has the form[7,20]

$$h=a_2\,\theta(1-\theta^2)r^{\beta+\gamma}, \tag{28a}$$

$$t=(1-b_2^2\theta^2)r, \tag{28b}$$

$$m=m(\theta)r^{\beta}, \tag{28c}$$

where

$$m(\theta)=k_2\,\theta(1+c\,\theta^2), \tag{28d}$$

$$b_2^2=3/(3-2\beta), \tag{28e}$$

$$c=(2\beta+2\gamma-3)/(3-2\beta). \tag{28f}$$

Here the parameter r is a direct measure of χ. The equation PE2 has not been previously applied to EuO. We obtained the fitting coefficients

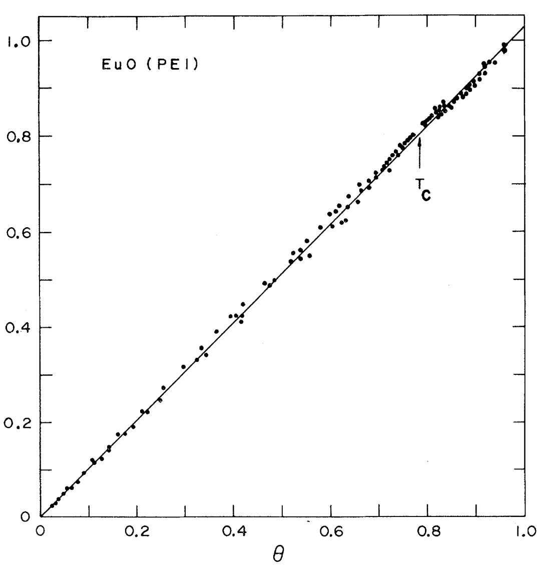

FIG. 4. Comparison between data and parametric equation PE1. The line is Eq. (20d).

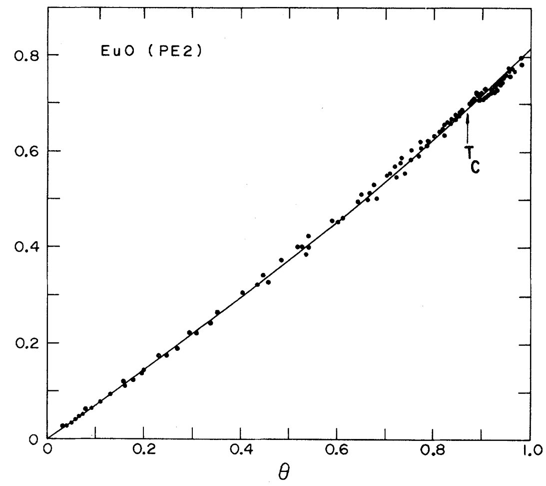

FIG. 5. Comparison between data and parametric equation PE2. The line is Eq. (28d).

$$a_2=1.78, \tag{29}$$

$$k_2=0.718. \tag{30}$$

The quality of the fit, as illustrated in Fig. 5, is quite satisfactory and is slightly better than that of PE1. The critical amplitudes according to PE2 are

$$B=k_2(1+c)(b_2^2-1)^{-\beta}, \tag{31}$$

$$\Gamma=k_2/a_2, \tag{32}$$

giving $B=1.22$ and $\Gamma=0.404$, again consistent with earlier results.

V. COMPARISON WITH OTHER EXPERIMENTS

The values of the critical exponents and amplitudes of EuO obtained in this and other experiments are summarized in Table I. It can be seen there is good agreement among the values of β measured by different techniques. Our result for γ, however, confirms the apparent pattern that the values of γ from static measurements agree with one another but disagrees with that obtained by neutron scattering. The origin of this discrepancy is unclear, but our corroboration of other static experiments lends some weight to the suggestion[24,25] that the expression of $\chi(q)$ used in the extrapolation of the neutron-scattering data at finite scattering wave vector q might not be appropriate in the presence of dipolar interactions. It should be pointed out that since the neutron scattering results do agree with theoretical predictions (see Sec. VI), there is always the possibility that static measurements are affected by dipolar interactions in a pathological way whereas the finite-q measurements are less sensitive.

Kornblit and Ahlers[6] have measured the specific heat C_p of EuO near the Curie point. Their result

TABLE I. Critical parameters of EuO.

β	γ	B	Γ	Technique	Reference
0.370 ± 0.006	1.30 ± 0.02	1.10 ± 0.04	0.397 ± 0.012	Faraday rotation	this work
0.368 ± 0.005	1.29 ± 0.01	1.22	0.375	Vibrating-sample magnetometer	1
$0.385^{+0.008}_{-0.028}$	1.315 ± 0.015	$1.255^{+0.03}_{-0.10}$	0.360 ± 0.017	Vibrating-sample magnetometer	4
0.36 ± 0.01	1.387 ± 0.036	1.17 ± 0.03		Neutron scattering	2
0.38 ± 0.03		1.12		High-temperature-series expansion	21
	1.405 ± 0.02			High-temperature-series expansion	22
	$1.375^{+0.02}_{-0.01}$		0.385	High temperature-series expansion	23
0.380	1.365			ϵ expansion (short range)	24
0.381	1.372			ϵ expansion (dipolar)	24

of fitting the singular part of C_p to the power-law functions,

$$C_p = (A/\alpha)\, t^{-\alpha} \quad (t > 0)\ , \tag{33}$$

$$C_p = (A'/\alpha')\, (-t)^{-\alpha'} \quad (t < 0)\ , \tag{34}$$

depends sensitively on the form of the correction terms used. With nonsingular correction terms, they obtained $\alpha = \alpha' = -0.044 \pm 0.01$ and $A/A' = 1.22 \pm 0.06$, while with singular correction terms, the optimum parameters are $\alpha = \alpha' = -0.10 \pm 0.05$ and $A/A' = 1.51 \pm 0.2$. We can compare these results with the specific-heat behavior predicted from our magnetization data. Using the scaling relation

$$\alpha + 2\beta + \gamma = 2 \tag{35}$$

and our values of β and γ, we obtain

$$\alpha = -0.04 \pm 0.03. \tag{36}$$

Since our data are found to be describable by the parametric equations of state, the amplitude ratio A/A' can also be derived in terms of critical exponents only. According to PE1, for example, the ratio is given by [19]

$$\frac{A}{A'} = \frac{1}{4}\left(\frac{\gamma}{\beta}\right)^2 \left(\frac{2\beta(\gamma-1)}{(1-2\beta)\gamma}\right)^{2\beta+\gamma}. \tag{37}$$

Our magnetization results therefore imply

$$A/A' = 1.31 \pm 0.05\ . \tag{38}$$

The parameters in Eqs. (36) and (38) are in excellent agreement with the specific-heat data of Kornblit and Ahlers analyzed using nonsingular correction terms, although they are also barely within the rather large error range of the results with singular correction terms.[6] Since the scaling equation of state we have obtained experimentally provides information on only the leading power-law behavior of C_p and not on the correction terms, a direct comparison between PE1 and the specific-heat data cannot be made at present. Specific-heat measurements in the presence of a magnetic field, which would enable correction-term effects to be eliminated, should be performed.

VI. COMPARISON WITH THEORY

Table I also contains the results of classical nearest-neighbor Heisenberg-model calculations. The values of the parmeters β, B, and Γ are in good agreement with our results. Our value of γ, however, is significantly less than the theoretical predictions. One possible explanation that has been explored is the effect of the appreciable next-nearest-neighbor interaction in EuO.[1,26] It is difficult to test this question conclusively on the basis of high-temperature-series calculations, but such a dependence of the exponent on the extent of the finite-range interaction would violate the concept of universality.[27] Another consideration is the role played by magnetic-dipole-dipole interactions. Renormalization-group calculations[24] have been made to second order in $\epsilon = 4 - d$, where d is the spatial dimensionality. A crossover behavior is predicted around $t = 0.05$, with the exponent γ changing from 1.365 to 1.372. We find that our susceptibility data can be accurately described by a single exponent, with no evidence of any crossover behavior. In any case, the expected change in γ is smaller than the accuracy of the experiment. It has been suggested that since the crossover temperature is in the middle of the experimental range, the constant amplitude of the susceptibility should be replaced by a scaling function,[24] but it is inconceivable how this can explain why the value of γ we obtained is smaller than both the theoretical short-range and dipolar values.

Finally, we have considered the effect of including corrections to scaling in our analysis of γ. It has been suggested that such corrections would add a factor of the form $(1+Ct^x)$ to all the power laws.[28,29] Assuming the existence of this factor in analyzing their specific-heat data, Kornblit and Ahlers estimated that $x=0.56\pm0.20$ and $C=-0.2\pm0.2$ for EuO.[6] Using these estimates and fitting the susceptibility data to Eq. (9) modified with the correction factor, we find the resultant γ to *decrease* by less than 0.02. Corrections to scaling, therefore, cannot account for the discrepancy with theory.

VII. SUMMARY

We have reported Faraday-rotation measurements on EuO near the Curie point. The results, after nonlinear corrections, yield critical exponents $\beta=0.370\pm0.006$ and $\gamma=1.30\pm0.02$. All the data can be scaled using these exponents, and are also consistent with two different parametric equations of state, the relevant coefficients of which are also presented. The result for γ agrees with other static experiments, but disagrees with neutron-scattering measurements and with theory. Our data predict specific-heat exponents and amplitudes which are consistent with experimental results analysed with nonsingular correction terms.

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*Present address: Department of Physics, University of Illinois, Urbana, Ill. 61801.

‡Present address: Department of Physics and Astronomy, State University of New York at Buffalo, Buffalo, N. Y. 14214.

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